Bioindicator studies in Nordic waters

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"Bioindicator Studies in Nordic waters"

Edited by Asker Aarkrog
Risø National Laboratory
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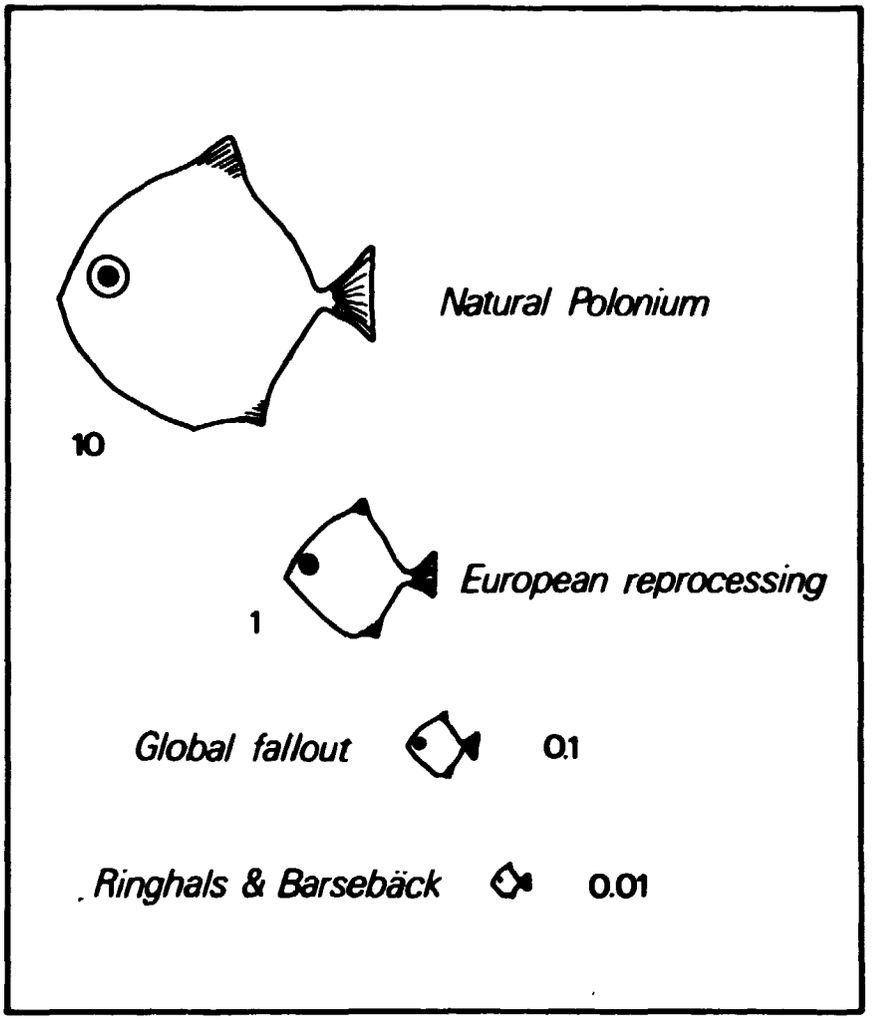

The fish symbolize the magnitude of the dose shown in man Sv in 1984 and should consequently be considered as spheres.

In the years to come the contribution from European reprocessing is expected to decrease as the discharges from Sellafield are reduced significantly.

The dose from global fallout is expected to decrease also, but more slowly.

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ABSTRACT

This project describes the application of bioindicator systems intended for the measurement of the low level radioactive contamination around nuclear installations. The system has been applied around the Swedish and Finnish nuclear power plants and has, furthermore, been used in a study of the dispersion of the effluents from the British nuclear reprocessing plant, Sellafield. The doses to man from these installations have been calculated and compared with the natural background radiation received from the consumption of marine fish.

INIS-descriptors:

AMERICIUM ISOTOPES; AQUATIC ECOSYSTEMS; BALTIC SEA; BARSEBAECK REACTOR; BIOINDICATORS; CESIUM 134; CESIUM 137; COBALT 58; COBALT 60; DENMARK; DOSES; ENVIRONMENT; FALLOUT; FAROE ISLANDS; FINLAND; FISHES; FORSMARK REACTOR ; GREENLAND; ICELAND; LOVIISA REACTOR; MAN; MANGANESE 54; MODELS; MUSSELS; NORTH ATLANTIC; NORWAY, OLKILUOTO REACTOR; OSKARSHAMN REACTOR; PLUTONIUM ISOTOPES; RADIOACTIVITY; RINGHALS REACTOR; SEAWATER; SEAWEEDS; SEDIMENTS; SELLAFIELD REPROCESSING PLANT; STRONTIUM 90; SVALBARD; SWEDEN; TECHNETIUM 99; TRANSFER FACTORS; ZINC 65;

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SUMMARY

Radioactive discharges from nuclear installations are usually small. The environmental concentrations of the radionuclides discharged are consequently often difficult to measure. However, certain organisms, so-called bioindicators, have the ability to accumulate radionuclides (and other substances). The evaluation of environmental levels thus becomes feasible by measurements on bioindicators.

This project has identified and calibrated two important marine bioindicators. The blue mussel: Mytilus edulis and the brown algae, bladder wrack: Fucus vesiculosus. These organisms are available from most parts of the northern North Atlantic region. They show biomagnification factors typically on the order of 100-10 000 for various radionuclides, i.e. 1 kg dry weight of the bioindicator contains as much activity as 0.1-10 m^3 of the water, in which the organism lives.

By means of bioindicators the radioactive contamination of the marine environment around the Nordic countries has been described. The important sources of this contamination are fallout from nuclear weapons testing in the atmosphere during the 1960s, waterborne discharges of radionuclides from the British nuclear reprocessing plant, Sellafield (formerly Windscale), and liquid effluents from nuclear power plants in Sweden, Finland and other countries in Northern Europe.

The radioactive contamination from a point source, e.g. a nuclear power plant, is approximately inversely proportional to the distance or squared distance from the plant. These distance relationships have been demonstrated by means of bioindicators surrounding the Nordic power plants.

The discharges from Sellafield have been traced by Fucus vesiculosus from the Irish Sea, along the Norwegian West coast to Svalbard, down along East Greenland and further up along West Greenland, i.e. over a distance of several thousand kilometers. Sellafield discharges also enter the Baltic Sea; the transit time from the Irish Sea being 4 years and the activity being diluted by a factor of approximately 100.

Transfer factors between discharges from nuclear installations and bioindicators at various distances from the installations have been estimated. By means of these transfer factors and from measurements of the bioindicators, it is possible to estimate the composition and amount of unreported discharges. Bioindicator systems may thus be applied by the authorities in their control of nuclear installations.

From the present studies it has been possible to estimate the mean residence time of water in the Danish straits to 0.2 years. From this information and from data on fish catch and consumption, the doses to man from waterborne discharges to the Danish straits have been calculated as 0.01 man Sv from 1 years operation of Barsebäck and Ringhals. This is approximately 1% of the dose received from Sellafield, and 1 o/oo of the dose received from naturally occurring ^{210}Po in the fish catch. The radioactive fallout from nuclear weapon tests in the sixties gives af collective dose of appriximately 0.1 man Sv from the fish consumption mentioned.

The accumulation of radionuclides in marine organisms depends upon various environmental factors, e.g. salinity and temperature. The waters around the Nordic countries vary from nearly 0 o/oo salinity up to 35 o/oo. Hence the bioindicator systems have to be calibrated in the environment where they should be applied. Laboratory experiments have elucidated the importance of the various environmental factors. Low salinity (or low temperature) often enhances the biomagnification factors in bioindicators.

A model which describes the seasonal variation in the uptake of radionuclides by Fucus has been set up. This model is based upon a combination of laboratory experiments and field observations. The predictions made from the model agree well with actual observations.

Another model based upon current velocity data is described in the appendix. This model calculates the dispersion and dilution of waterborne contaminants in the area north of Barsebäck. The results obtained by the model have been verified by bioindicator measurements.

The need for better radiochemical methods have resulted in significant improvements within low level radiochemistry. Special equipment and apparatus for laboratory and field studies have also been developed.

During the project 74 publications have been produced: 3 Ph.D. theses, 9 articles in refereed scientific journals, 19 presentations at international meetings, 28 at Nordic meetings and 25 reports with international distribution.

In summary, an environmental monitoring system has been developed for the surveillance and control of liquid discharges from nuclear installations. The system is applicable for normal operation as well as in the case of accidental releases. As environmental and operational conditions at nuclear installations may change in time, it is important that the system is permanently updated and revised in order to optimize its performances. It is therefore recommended that bioindicator studies, in field as well as in laboratory, should be continued.

SAMMENFATNING (in Danish)

Radioaktive udslip fra nucleare anlæg er sædvanligvis små. Som følge heraf er det ofte vanskeligt at måle koncentrationerne af de udledte radionuklider i miljøet. Imidlertid har visse organismer, de såkaldte bioindikatorer, evnen til at akkumulere radionuklider (og andre stoffer). Herved bliver det muligt at bestemme niveauerne i miljøet ved at måle på sådanne bioindikatorer.

I det foreliggende projekt har man identificeret og kalibreret to vigtige marine bioindikatorsystemer. Blåmuslingen: Mytilus edulis og brunalgen, blæretang: Fucus vesiculosus. Disse organismer findes næsten overalt i den nordlige del af det nordatlantiske område. De udviser akkumuleringsfaktorer typisk i størrelsesordenen 100-10 000. D.v.s. 1 kg tørvægt af en bioindikator indeholder lige så meget af det radioaktive stof, som 0,1-10 m^3 af det vand organismen lever i.

Ved hjælp af bioindikatorer er den radioaktive forurening af det marine miljø omkring de nordiske lande blevet beskrevet. Hovedkilderne til denne forurening er radioaktivt nedfald fra kernevåbenforsøg i atmosfæren i 1960'erne, vandbårne udledninger af radioaktive stoffer fra det engelske oparbejdningsanlæg for reaktorbrændsel, Sellafield (tidligere Windscale) og vandbårne udledninger fra kernekraftværker i Sverige, Finland og andre lande i Nordeuropa.

Den radioaktive forurening fra en punktkilde, f.eks. et kernekraftværk er tilnærmelsesvis omvendt proportionalt med afstanden eller kvadratet på afstanden fra anlægget. Disse afstandsrelationer er blevet påvist ved hjælp af bioindikatorer indsamlet omkring de nordiske kernekraftværker.

Udledningerne fra Sellafield er blevet sporet ved hjælp af Fucus vesiculosus fra det Irske Hav, langs den norske vestkyst, op til Svalbard, ned langs Øst Grønland og videre op langs Vest

Grønland. D.v.s. over en afstand af adskillige tusinde kilometer. Sellafieldudslippene når også ind i Østersøen; transporttiden fra det Irske Hav er hertil 4 år og aktiviteten bliver fortyndet med ca. en faktor 100.

Overføringsfaktorer fra udslip fra nukleare anlæg til bioindikatorer i forskellige afstande fra anlæggene er blevet bestemt.

Ved hjælp af disse overføringsfaktorer og udfra målinger på bioindikatorer, er det muligt at bestemme sammensætning og mængden af urapporterede udslip. Bioindikatorsystemer kan således benyttes af myndighederne i kontrollen med nukleare installationer.

Fra de foreliggende bioindikatorundersøgelser har det været muligt at bestemme middel opholdstiden for vandet i de danske stræder til 0,2 år. Fra denne oplysnig og udfra statistikker over fiskefangst og fiskekonsum er den kollektive dosis til mennesker beregnet. Væskebårne udslip til de danske stræder fra et års drift af Barsebäck og Ringhals resulterer i en dosis på ca. 0,01 manSv. Dette er ca. 1 % af den dosis man får fra Sellafieldforureningen i fisk fanget i Kattegat og ca. 1 o/oo af den dosis man modtager fra naturligt forekommende ^{210}Po i denne fiskefangst. Det radioaktive nedfald fra kernevåbenforsøg i tredserne giver en kollektiv dosis på ca. 0,1 manSv fra det ovennævnte fiskekonsum.

Akkumuleringen af radionuklider i marine organismer afhænger af miljøfaktorer, som f.eks. salinitet og temperatur. Vandene omkring de nordiske lande varierer fra næsten 0 op til 35 o/oo i salinitet. Bioindikatorsystemerne må derfor kalibreres i det miljø, hvor de skal benyttes. De forskellige miljøfaktorers betydning er blevet belyst gennem laboratorieforsøg. Lav salinitet (eller lave temperaturer) vil ofte øge akkumuleringsfaktorerne for bioindikatorer.

En model der beskriver årstidsvariationen i optagelsen af radionuklider i Fucus er blevet udviklet. Denne model er baseret på en kombination af laboratorieeksperimenter og feltobservationer.

Modellens forudsigelser er i god overensstemmelse med de faktiske iagttagelser.

En anden model baseret på strømmålinger er beskrevet i appendix. Denne model redegør for spredningen og fortyndingen af vandbårne stoffer i området nord for Barsebäck. Modellens resultater er blevet verificeret med bioindikatormålinger.

Behovet for bedre radiokemiske metoder har resulteret i væsentlige forbedringer indenfor "low level" radiokemien. Under projektet er der blevet udviklet specielt apparatur og udstyr til laboratorie- og feltarbejdet.

Projektet har affødt 74 publikationer: 3 Ph.D. afhandlinger, 9 artikler i internationale videnskabelige tidsskrifter, 19 foredrag ved internationale møder, 28 ved nordiske møder og 25 rapporter med international udbredelse.

Dette projekt har udviklet et miljøovervågningssystem for vandbårne radioaktive udledninger fra nukleare anlæg. Systemet kan anvendes ved rutinemæssige udledninger, såvel som i tilfælde af uheldsudslip. Da miljø - såvel som driftsmæssige forhold - kan ændres med tiden omkring nukleare anlæg er det vigtigt, at et bioindikatorsystem til stadighed revideres og ájourføres for at opnå den størst mulige nyttevirkning og effektivitet. Det må derfor anbefales, at man også fremover viderefører bioindikatorundersøgelserne i de nordiske lande. Dette vil tillige betyde en yderligere styrkelse af det nordiske samarbejde indenfor kernesikkerhedsforskningen som er blevet etableret under dette projekt.

1. INTRODUCTION

1.1. General

The radioactive pollution from nuclear installations is usually so small, that the emitted radionuclides in air, water and soil are difficult to measure. One might then just accept this and conclude that there consequently has been no radioactive contamination and thus no dose given to the population from the installation. Such a conclusion would, however, be false and it also would not be in accordance with the International Commission for Radiological Protection. ICRP conservatively assumes that even the smallest amounts of ionizing radiation has a deleterious effect, which might result into economic consequences for the society. ICRP recommends that the operation of nuclear facilities is organized in such a way that the harmful effects are outweighed by the benefits to the society from the exploration of nuclear technique. Hence, in order to perform a cost-benefit analysis it is necessary to have as reliable information as possible on the doses from routine discharges. If too pessimistic assumptions are made the doses will be overestimated and the cost-benefit analysis will rely on an unrealistic assumption.

1.2. Bioindicators

A bioindicator is an organism whose contents of radionuclides or stable metals is used to indicate the level of radioactive or trace metal pollution in the environment in which the organism is living.

Bioindicators may thus be used for detecting and indirectly measuring the radionuclide concentrations in the environment around nuclear facilities. With proper conversion factors the concentrations in bioindicators may be applied for a calculation of the levels in foodstuffs produced in the same area as the bioindicators. From the concentrations in diet it is possible to calculate the intake of the various radionuclides and hence of the doses to man.

Several reasons for using bioindicators instead of water samples can be noted: 1) An integration of fluctuating levels is made over a period of time, 2) due to accumulation, the limit of detection is lowered, 3) an estimate can be made of transfer to humans via edible organisms, and 4) there is the highest sensitivity for the most biologically available physicochemical species, i.e., if a radionuclide exists on a certain form, which is not accumulated in the bioindicator, it is, ideally, neither accumulated in species used for human food, and is thus less important than if it had existed in a highly available form.

1.3. Purpose of the NKA bioindicator project

This project was initiated in 1978 under the auspices of the Nordic Council of Ministers. The purpose was:

a) to identify bioindicator systems, which were suited for monitoring radioactive contamination in the Nordic environment;
b) by means of these bioindicators to describe the radioactive contamination of the Nordic Waters, including the variation with time and location;
c) to calculate transfer factors from discharges from Nordic nuclear installations to these bioindicators;
d) to calculate the doses from the consumption of marine products contaminated by radionuclides from nuclear installations.

e) By laboratory and field experiments to identify the significance of various environmental factors, e.g. salinity and temperature for the uptake of radionuclides by the bioindicators.
f) To set up dispersion models for the radioactive contamination of the marine environment based upon the data acquired during this project.

2. OUTLINE OF THE PROJECT

The NKA bioindicator project has been divided into a number of subprojects:

- The Panscandinavian Fucus Project. (2.1.)
- Calibration of the recipients around nuclear facilities. (2.2.)
- Experimental Fucus and Mytilus experiments. (2.3.)
- The Baltic Gauss Project. (2.4.)
- Models for dispersion of radioactive contamination in the marine environment. (2.5.)
- Development of methods and instruments. (2.6.)

2.1. The Panscandinavian Fucus Project has comprised collection of fucoids along the coasts of the Nordic countries, the Faroe Islands, Greenland, Svalbard and Iceland included. The samples have been analysed for ^{60}Co, ^{90}Sr, ^{99}Tc, ^{134}Cs, ^{137}Cs, $^{239,240}Pu$ and ^{241}Am. (Institutions: IFE, SIS, N, LUND, GOTU, RISØ, STUK (cf. 2.7 on page 12-13).

2.2. The calibration of recipient project was performed at the nuclear power plants: Loviisa, Olkiluoto, Forsmark, Oskarshamn, Ringhals and Barsebäck. Samples of Fucus, Mytilus, sediments and seawater have been collected and compared with discharge data. The samples have been analysed by γ-spectroscopy of γ-emitters (Institutions: LUND, RISØ, STUK, SNV).

2.3. The experimental Fucus and mytilus studies have been carried out in the health physics laboratory at Risø and at the Biotest lake at Forsmark. Furthermore, field studies with Fucus were carried out at Ringhals, Barsebäck and Risø. The experiments were carried out with ^{60}Co, ^{65}Zn, $^{95}Tc^{m}$, ^{134}Cs, ^{144}Ce, ^{155}Eu, Pu, Am, Cm and Np. (Institutions: RISØ, SNV.)

2.4. The Baltic Gauss Project took place on two cruises with the vessel F/S Gauss from the German Hydrographic Institute in Hamburg. Samples of seawater, sediments and benthic biota were collected and have been analysed for γ-emitters (^{134}Cs, ^{137}Cs) and transuranics (Pu, Am). (Institutions: DHI (Hamburg), LUND, STU, STUK, RISØ.)

2.5. The projects with dispersion models have been carried out around Barsebäck and Ringhals. Current measurements have been made. Furthermore, the measurement of ^{60}Co in Fucus and the ^{137}Cs measurements of seawater have been used in this project. (Institutions: RISØ, LUND, STUK, SNV.)

2.6. Development of methods and instruments have comprised radiochemical methods for ^{60}Co, ^{63}Ni, ^{99}Tc and radiocesium. Furthermore, instruments for low-level β-measurements, algae feeding and continuous water sampling have been developed. (Institutions: LUND, RISØ).

2.7. Participants in the project

Denmark: Risø National Laboratory (RISØ) (Asker Aarkrog projectleader), Henning Dahlgaard, Søren Boelskifte, Lars Bøtter-Jensen and Annelise Meide.

Finland: Finnish Centre for Radiation and Nuclear Safety (STUK) (Erkki Ilus, Jaakko Ojala, Kirsti-Liisa Sjöblom and Kristiina Tuomainen).

Norway: Institute for Energy Technology (IFE) (Gordon Christenson). National Institute of Radiation Hygiene (SIS, N) (Torolf Bertelsen).

Sweden: Lund University (LUND) (Elis Holm, Lars Hallstadius, Simon Duniec, Björn Bjurman, Robert Hedvall, Ulf Nilsson, Juan Rioseco, Lena Carlsson, Bengt Erlandsson). National Swedish Environmental Protection Board (SNV) (Manuela Notter, Georg Neumann, Magnus Edgren). Gothenburg University (GOTU) (Sören Mattsson). Studsvik Energiteknik AB (STU) (Per-Olof Agnedal, Svante Lampe).

3. RESULTS AND DISCUSSION

3.1. Development of methods and equipment

3.1.1. Analytical methods

During the project the need for new and better radiochemical methods has resulted in significant improvements within low level radiochemistry. We have thus developed analytical techniques for large volume (~ 0.2-2 m^3) seawater samples; methods for determining of transuranics (Pu, Am, Np), radiocesium, ^{60}Co and ^{99}Tc in water are briefly described below. We have furthermore developed methods for radiochemical analysis of ^{99}Tc and ^{63}Ni on biological samples, in particular seaweed.

3.1.1.1. Transuranium analysis on large seawater samples.

This method is based upon the classical procedure described by Talvitie (Analyt. Chem. 43, 1827-30 (1971)) and by Holm et al.

(Talanta 26, 791-794 (1979)). The samples of seawater are collected in 0.2 or 2 m^3 tanks and precipitated by NaOH (pH = 10) on board the ship in the presence of ^{242}Pu and ^{243}Am spikes. The precipitates are brought to the laboratory for further radiochemical processing. This procedure has also been used on airborne expeditions where large plastic bags (0.1 m^3) suspended under a phototripod were used instead of the tanks. (NKA References: 1.2, 1.3, 1.6, 1.7, 3.1, 3.8, 3.14, 4.9.)

3.1.1.2. Technetium-99 analysis. Technetium-99m ($t_{1/2}$ = 6.0h) is used as a radiochemical yield determinant. For large water samples Tc is reduced to the +IV oxidation state and co-precipitated with iron hydroxide. Technetium is extracted from H_2SO_4 solution with tributylphosphate and backtracted with NaOH solution. Technetium is electroplated from 2M NaOH onto stainless steel discs. The yield is determined by γ-spectroscopy using the 140 keV transition from $^{99}Tc^m$. After decay of $^{99}Tc^m$, ^{99}Tc is measured by an anticoincidence shielded G.M-gas flow counter. The radiochemical yield of 10-100g biological samples is 50-80%.
(NKA references: 1.8, 3.3, 3.8, 3.13, 3.14, 3.15, 3.16, 4.11, 5,19, 5.27.)

3.1.1.3. Cobalt-60 analysis. Seawater is collected in a tank (0.2-2 m^3); $Co(NO_3)_2$ is added as carrier and ^{57}Co as a radiochemical yield determinant. CoS is precipitated by adding Na_2S to the solution. The yield is 100% and the precipitate furthermore contains all the ^{65}Zn activity (if any) in the water. The precipitate is measured by Ge(Li) spectroscopy and ^{57}Co, ^{58}Co and ^{60}Co is determined.
(NKA references: 1.4, 2.1, 3.5, 4.10, 5.14, 5.22.)

3.1.1.4. Radiocesium determination on large seawater samples. Yamagata (Nature 200, 157 (1963) has shown how AMP (ammonium molybdophosphate) can be used for precipitation of radiocesium from seawater. Usually 50 g AMP has been used for 50 ℓ seawater. We have found that we recover 60-70% of the activity in 2 m^3 of seawater if we use 100 g AMP. In order to concentrate the activity, we dissolve the AMP in NaOH and reprecipitate it as Cs_2PtCl_6. By this procedure and by counting the samples for 1 week our limit of detection for ^{134}Cs becomes 0.01 Bq m^{-3}.

We have noticed that in the Baltic Sea we get lower yields than expected. This is not caused by the lower salinity, but probably to some organic complexing agent, which disturbs the reaction between AMP and the cesium in the seawater. (A. Münsberg & Ib Olsen: Precipitation of cesium in seawater, an optimization experiment (Internal report in Danish)).
(NKA references: 1.1, 1.3, 1.7, 3.2, 3.8, 3.9, 3.14, 4.7, 4.8, 4.9, 4.10, 4.11.)

3.1.1.5. Nickel-63 determination. This procedure is under development. Nickel-carrier is added to the sample. The sample is ashed and dissolved in aqua regia. Nickel is precipitated as a chelate with dimethylglyoxim. The nickel chelate is dissolved and passed through a Dowex 1 x 8 anion exchange column. The resin retains Co and Fe. The Ni-solution from the column is treated with Fe-scavenging, and finally $Ni(OH)_2$ is precipitated. The last step is an electrodeposition of the Ni on a silver disc. The chemical yield is determined by X-ray fluorescens. The sample is due to the low β-energy counted on a windowless GM-flow-counter.
(NKA references: 5.23, 5.26.)

3.1.2. Instruments

3.1.2.1. The beta multicounter system. The measurements of low level, low beta energy samples, such as ^{99}Tc and ^{63}Ni, demand advanced counting equipment. Risø has developed a GM-25-5 low-level beta multicounter system, which partly consists of a gas flow counter unit incorporating 5 individual G-M counter elements and a common guard counter. The guard counter reduces the cosmic background by using anticoincidence technique. The background is typically less than 0.2 cpm. The efficiency for e.g. electroplated ^{99}Tc is 42%. During the project, this counting system has been delivered to laboratories in Sweden, Monaco, Canada, Australia and Finland.
(NKA references: 1.8, 5.23, 5.26.)

3.1.2.2. A turbidity controller. In the bioaccumulation studies of mussels phytoplankton suspensions are applied as food to the animals. An apparatus called a turbidity controller has been designed to maintain the phytoplankton cell concentrations at a constant and very low level. The set up facilitates continuous control of animal condition by measuring the clearance rate.
(NKA references: 1.5, 2.2.)

3.1.2.3. Continuous water sampler. This set-up has been developed in order to measure the concentration factor from water to Fucus at the Swedish power plants Barsebäck and Ringhals. The water is pumped from the cooling water channel to a 400 ℓ container. The pump has a capacity of approximately 13 ℓ per day or 400 ℓ per month. At the end of each month a precipitation is made and the precipitate is measured on a Ge(Li) γ-counter.
(NKA references: 1.4, 2.1, 3.5, 5.22.)

3.1.3. Intercalibration

During the project there have been several intercomparison exercises. Intercalibration samples have been provided by the Nordic Laboratories as well as by the IAEA. In general, the analytical performances by the participating laboratories have been satisfactory. The relative standard deviation between laboratories were thus 26% for Pu analysis and 7% for ^{137}Cs.

We have also compared various sediment samplers in another Nordic exercise.
(NKA reference: 3.1, 3.2, 3.14, 4.5, 4.11.)

3.2. Field studies

3.2.1. The Panscandinavian Fucus project

This project has been a co-operative effort among 1) Institute for Energy Technology, Kjeller, Norway, 2) Finnish Centre for Radiation and Nuclear Safety, Helsinki, Finland, 3) University of Lund, Sweden, 4) University of Gothenburg, Sweden, and 5) Risø National Laboratory, Roskilde, Denmark.

The purpose has been to use fucoids, in particular Fucus vesiculosus, to describe the distribution of radionuclides in the marine environment of the North Atlantic region.
Figs. 3.2.1.1.-3.2.1.4. summarize the results.

Figure 3.2.1.1. shows that the ^{137}Cs concentrations are nearly constant within the Baltic area (8-10 Bq kg^{-1} dry weight). Lower concentrations are seen in south-west Jutland and the Netherlands (3-6 Bq kg^{-1}). Along the Norwegian west coast the concentrations decrease from 13 in the south to 6 in the north. At Svalbard and in East Greenland the levels are around 1-2 Bq kg^{-1} and in Iceland and west Greenland they are less than

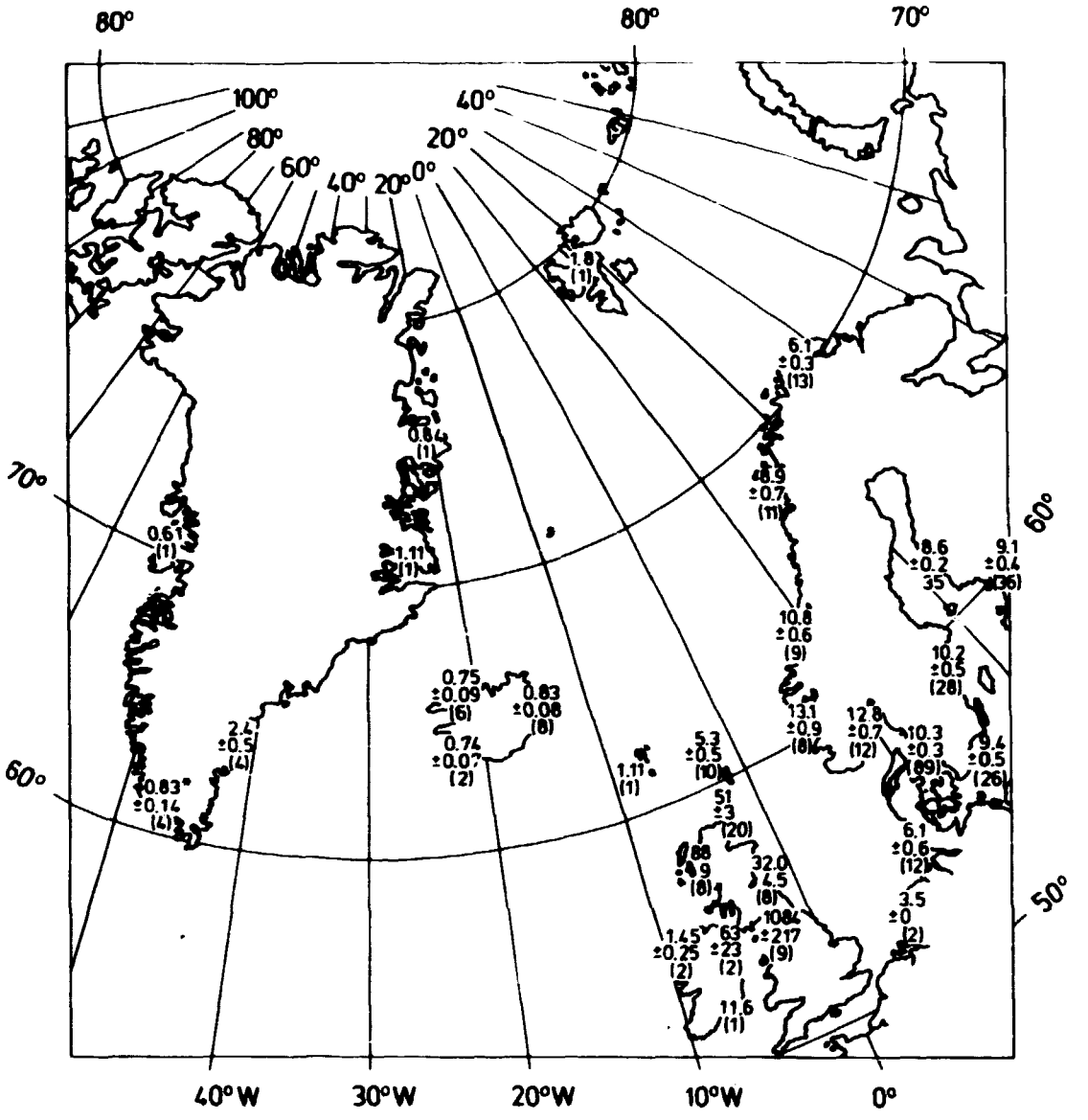

Fig. 3.2.1.1. Cesium-137 in Fucus in the Northern North Atlantic 1980-1983. (Bq kg^{-1} d.w. ± 1 SE (number of samples)).

1 Bq kg^{-1}. High concentrations (1000 Bq kg^{-1}) are found close to Sellafield. The concentrations on the Irish east coast are lower than those on the British west coast. We conclude that the ^{137}Cs contamination from Sellafield by means of fucoids has been traced along the British and Irish coastlines, to the Danish Straits and along the Norwegian coastline. From Sellafield to the Danish Straits the concentrations decrease by a factor of 100.

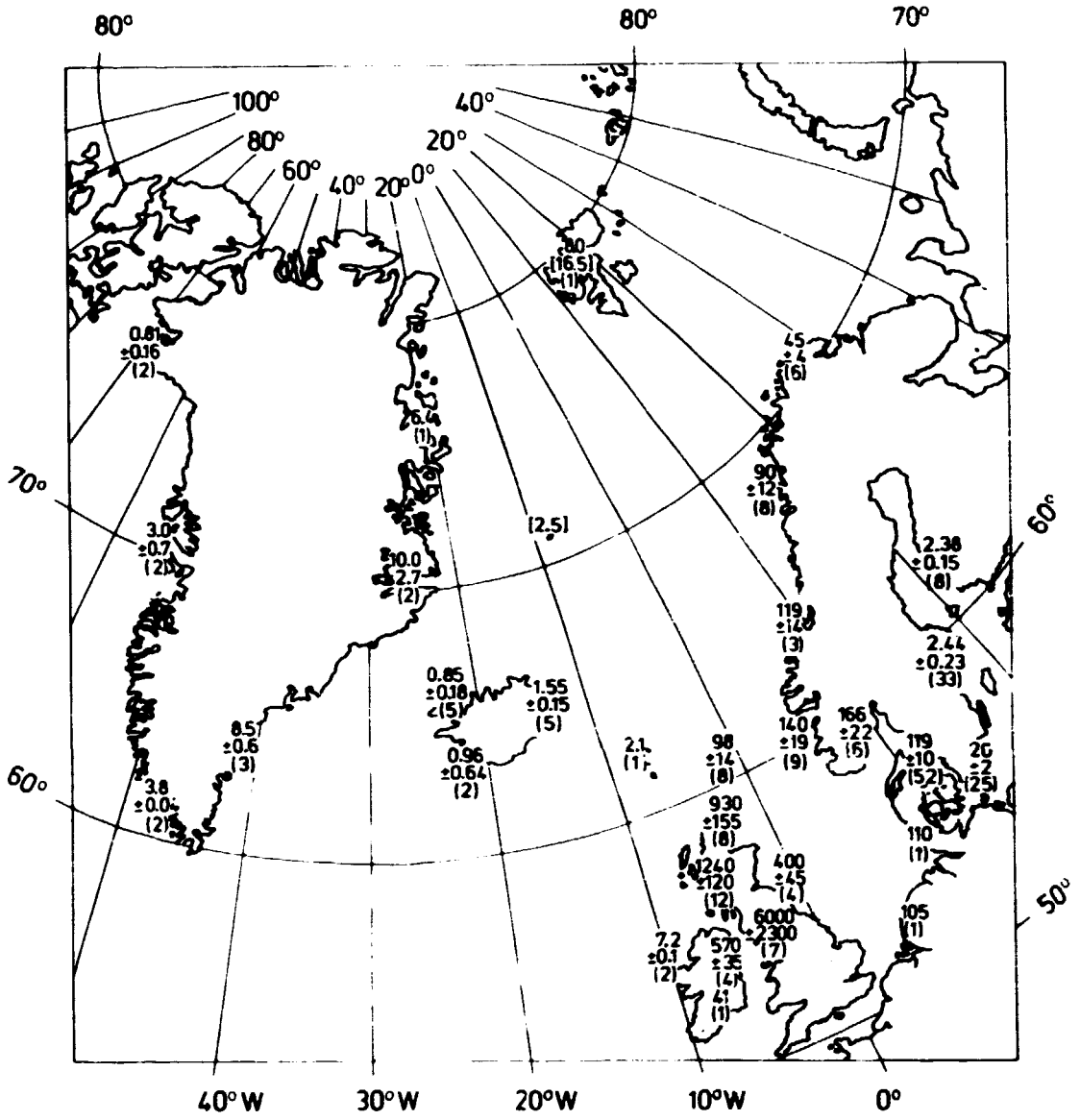

Fig. 3.2.1.2. Technetium-99 in Fucus in the Northern North Atlantic 1980-1983. (Bq kg^{-1} d.w. ± 1 SE (number of samples)).

Fig. 3.2.1.2. shows that ^{99}Tc follows the same pattern as ^{137}Cs, but the gradients for ^{99}Tc are in some cases more pronounced than those seen for ^{137}Cs. In the Baltic area the levels vary from 120 in the Cattegat to 2 in the Gulf of Botnia. Along the Norwegian west coast the levels decrease from 165 in the south to 45 in the north. Because of the lower fallout background we are able to trace ^{99}Tc from Sellafield to the Greenland west coast, i.e. over a distance of 7000-8000 km. The concentration range is 1: (10^3-10^4).

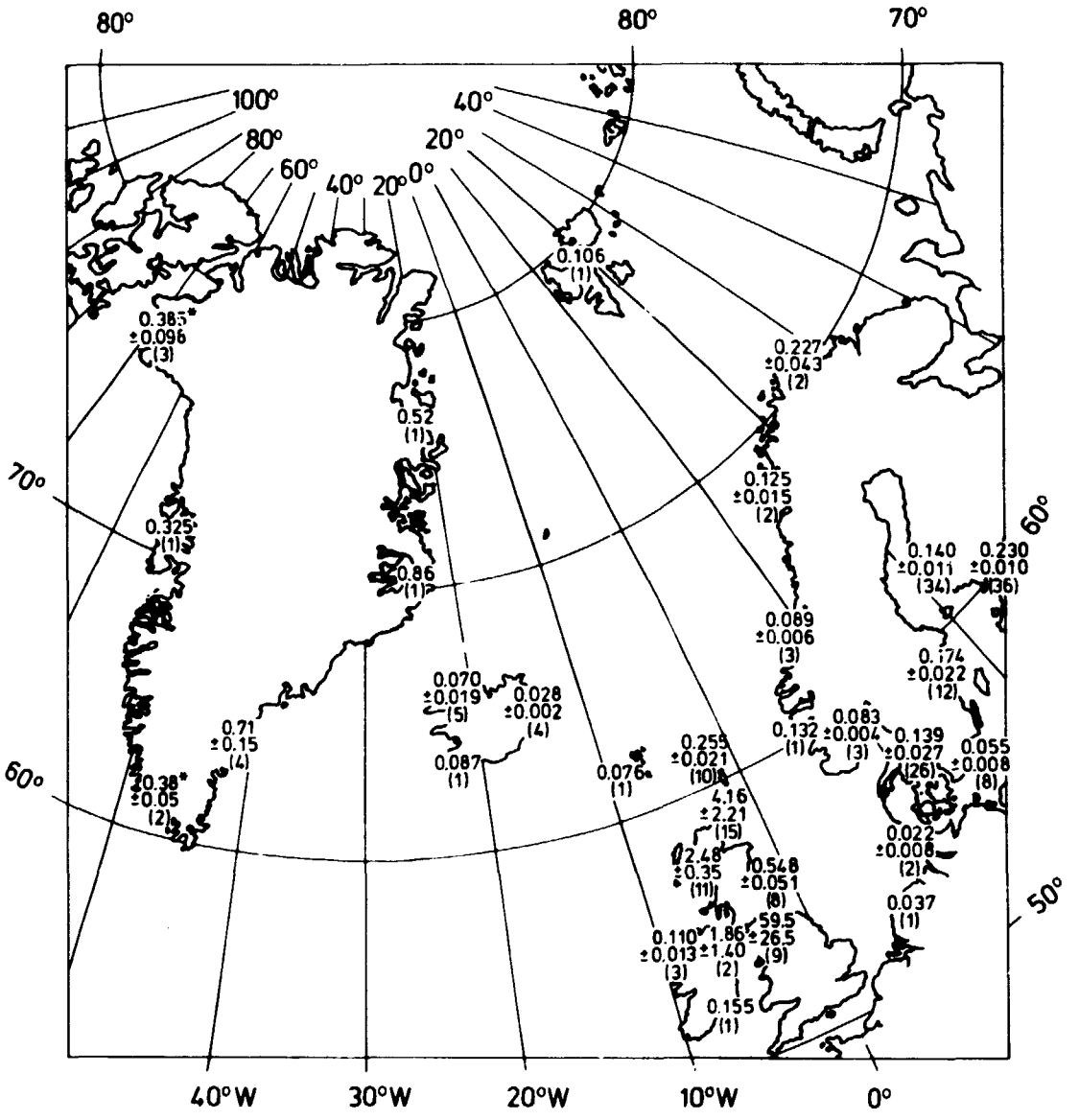

Fig. 3.2.1.3. Plutonium-239,240 in Fucus in the Northern North Atlantic 1980-1983. (Bq kg^{-1} d.w. ± 1 SE (number of samples)).

Plutonium 239,240 is also discharged from Sellafield (cf. Fig. 3.2.1.3.) but in this case enhanced levels are seen only in British and Irish coastal waters. The relatively high concentrations seen in cold waters (at Greenland) are probably due to the slow growth of the plants and hence to the accumulation of plutonium over long periods. Similar reasons may be found for the enhanced concentrations seen in the Gulf of Finland and in northern Norway. The lowest concentrations of Pu were found in south-western Jutland, The Netherlands and in eastern Iceland.

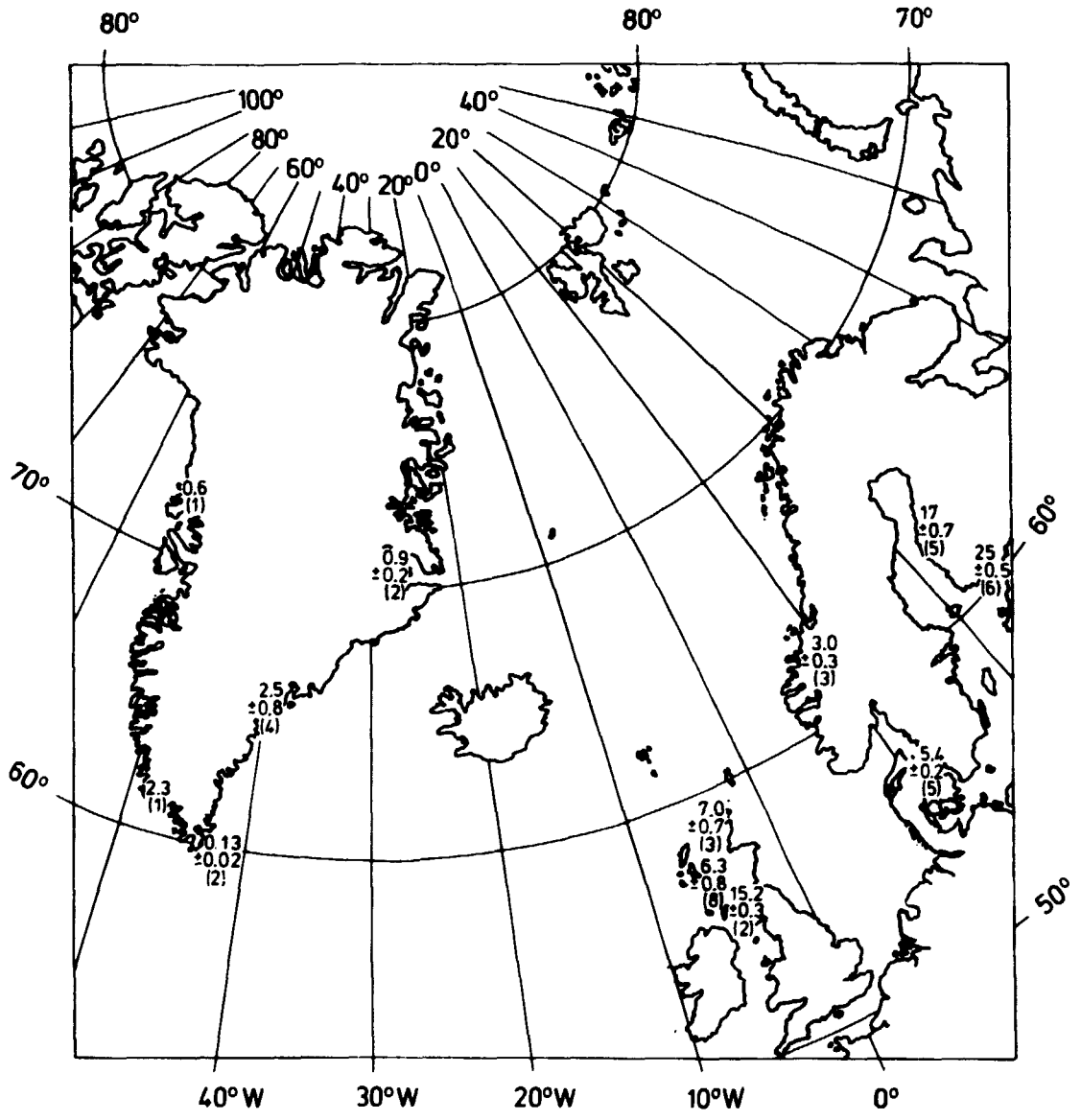

Fig. 3.2.1.4. Strontium-90 in Fucus in the Northern North Atlantic 1980-1983. (Bq kg^{-1} d.w. ± 1 SE (number of samples)).

Strontium-90 has been measured less extensively than the other nuclides (cf. Fig. 3.2.1.4). In this case the Gulfs of Finland and Bothnia show the highest concentrations, but samples collected in the Irish Sea were not analysed for ^{90}Sr. The levels along the Greenland coastline vary by a factor of 20 which is more than seen for the other nuclides in this study. We have no evident explanation for this phenomenon.

The time variation of ^{99}Tc in Fucus vesiculosus collected in the Danish Straits since 1967 is shown in Fig. 3.2.1.5. From the measurements of ^{137}Cs in seawater and in Swedish Fucus samples we know that the transit time from Sellafield to the Straits is four years. As the increase in ^{99}Tc began in 1974 we conclude that enhanced discharges of ^{99}Tc began in 1970.

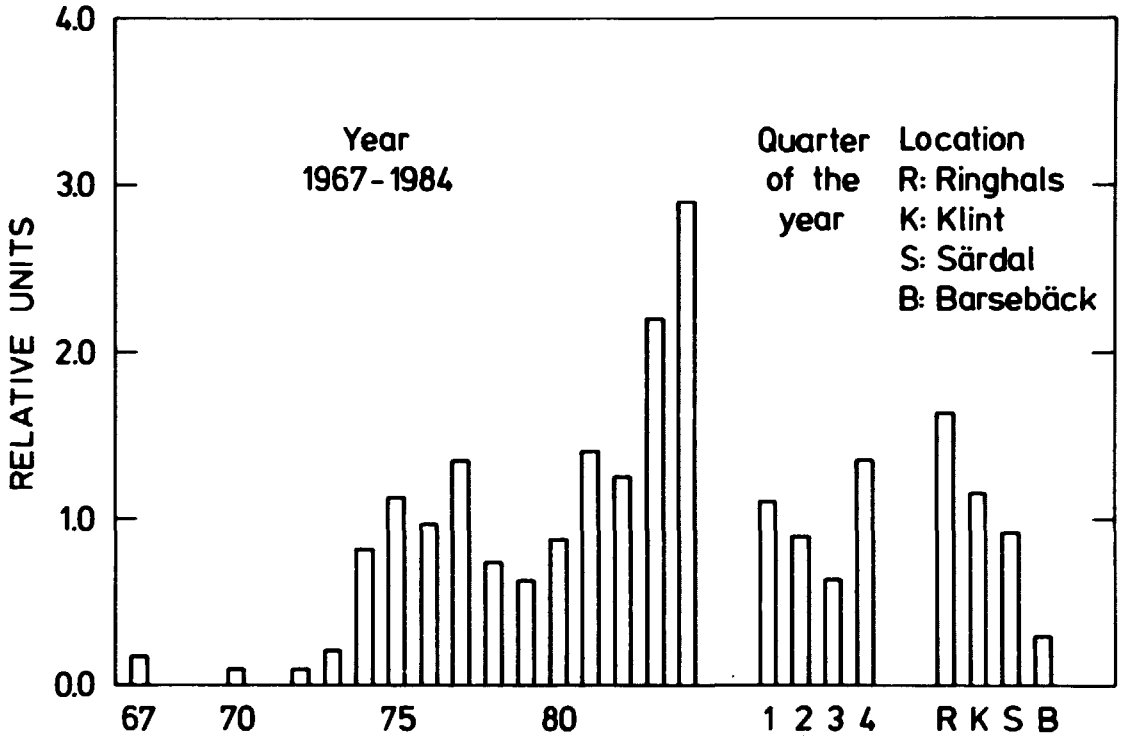

Fig. 3.2.1.5. Time variation of ^{99}Tc in Fucus in the Danish Straits 1967-1984.

The Fucus project has also been used for a determination of concentration factors between seawater and Fucus in the various parts of the North Atlantic region Figure 3.2.1.6. and 3.2.1.7. summarize the results for $^{239,240}Pu$ and ^{137}Cs. In general, low salinities show in general higher concentration factors than high salinities, and Polar waters show higher concentration factors for Pu than temperate waters. For temperate ocean water the concentration factor for ^{137}Cs is 2×10^2 and for $^{239,240}Pu$ 10^4 and in case of ^{90}Sr we found 10^2.

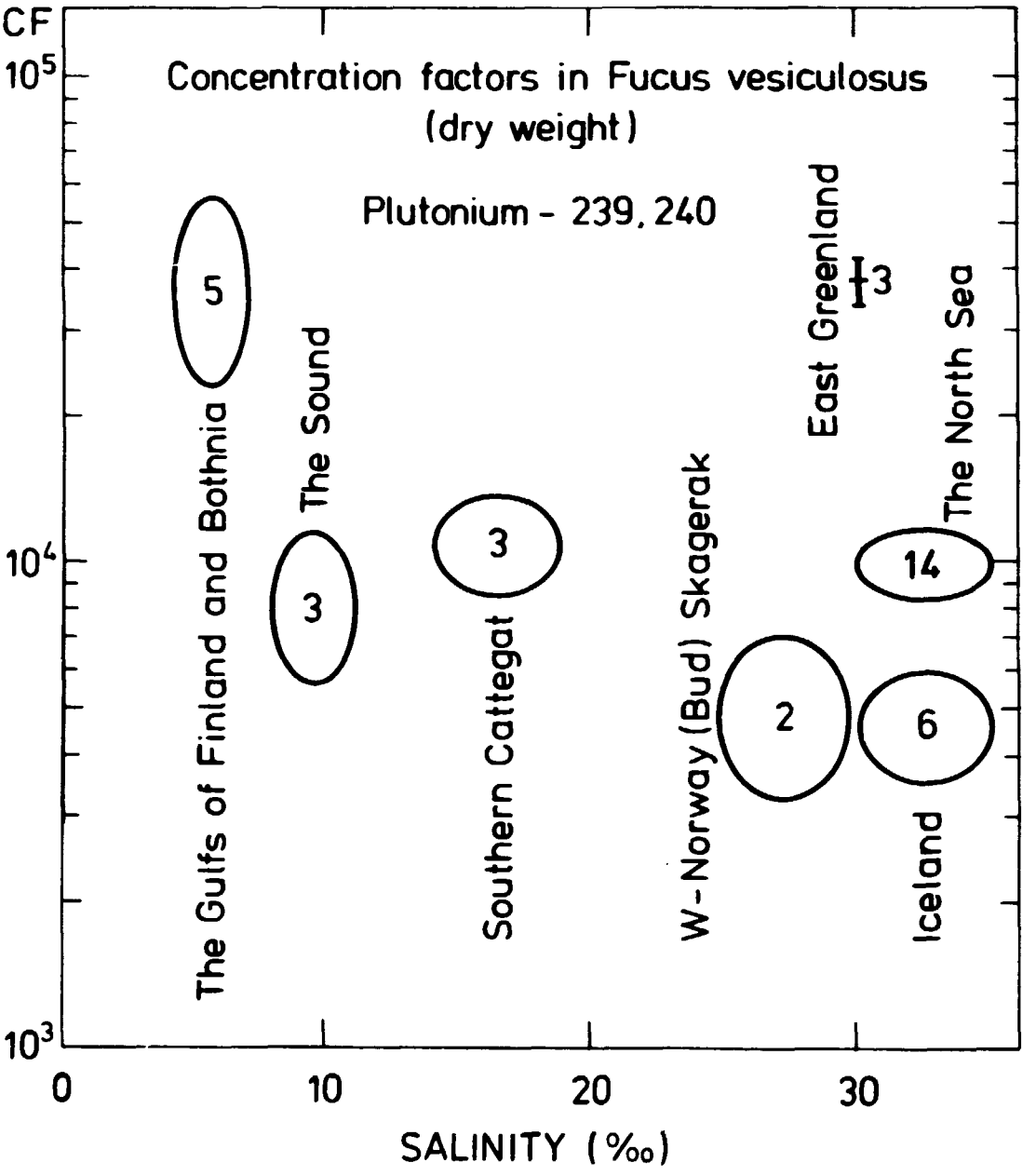

Fig. 3.2.1.6. Concentration factor Fucus/sea water for $^{239,240}Pu$ in the Northern North Atlantic (number of determinations and 1 SE indicated).

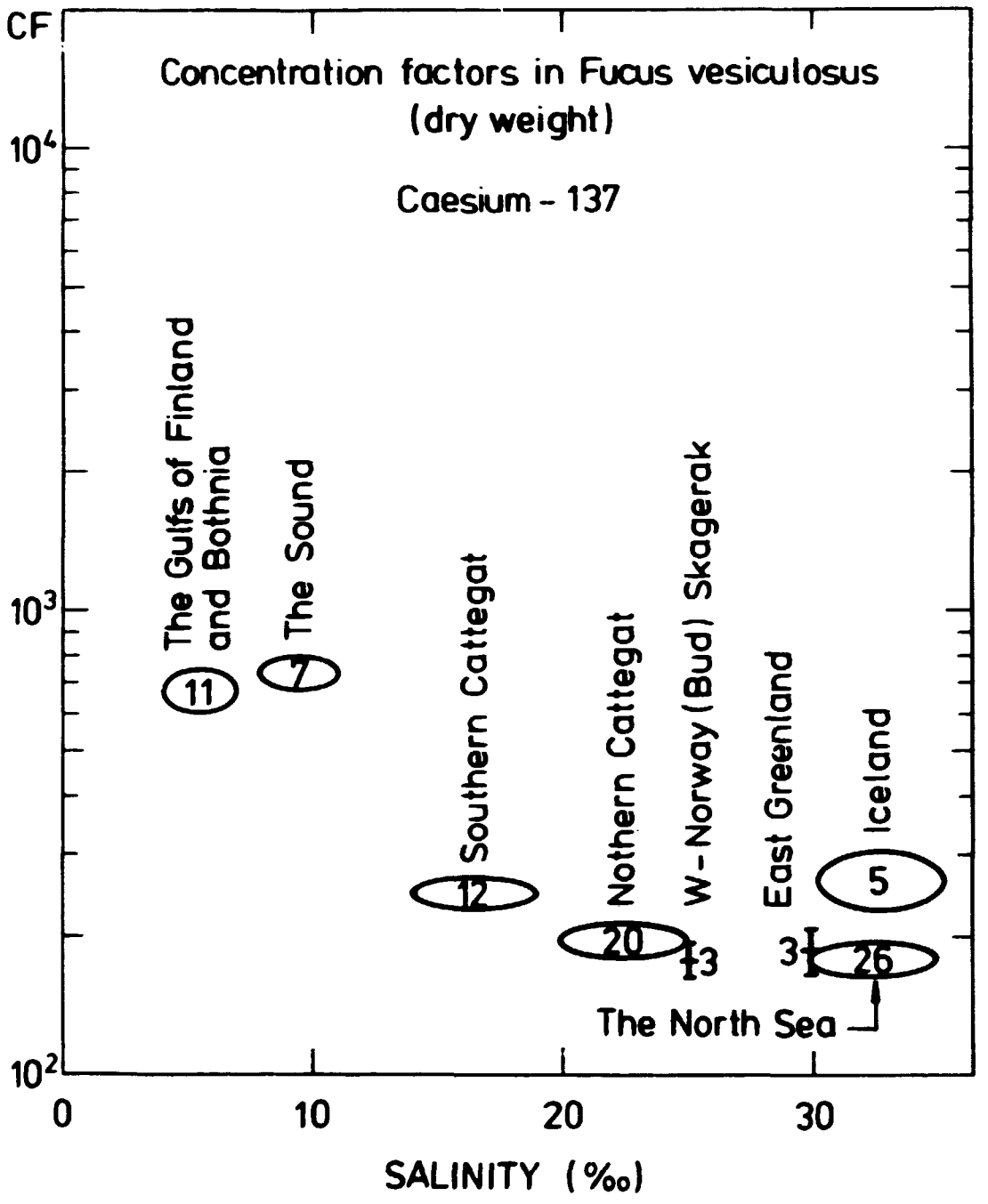

Fig. 3.2.1.7. Concentration factor Fucus/sea water for ^{137}Cs in the Northern North Atlantic (number of determinations and 1 SE indicated).

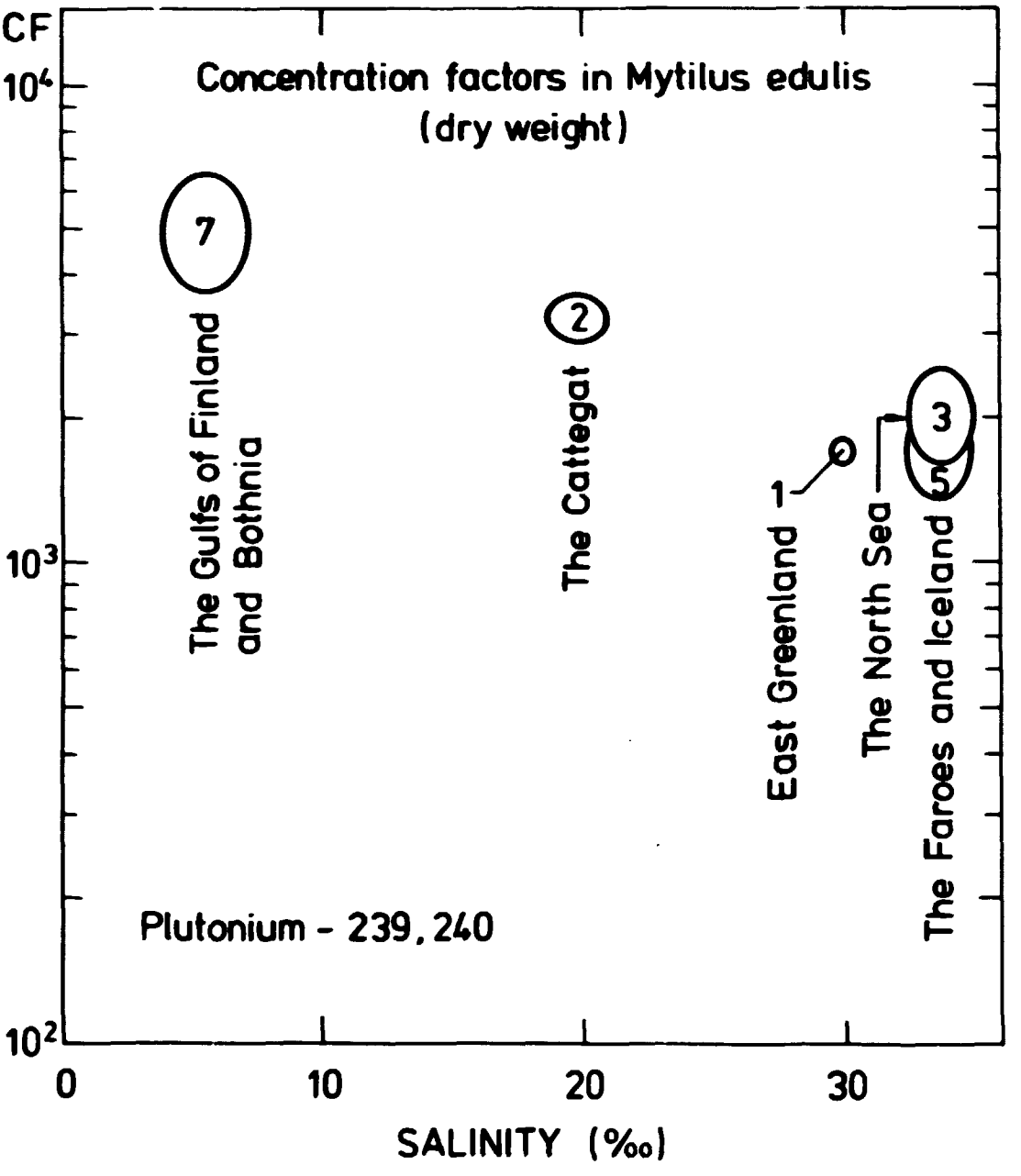

Fig. 3.2.1.8. Concentration factor Mytilus/sea water for $^{239,240}Pu$ in the Northern North Atlantic (number of determinations and 1 SE indicated).

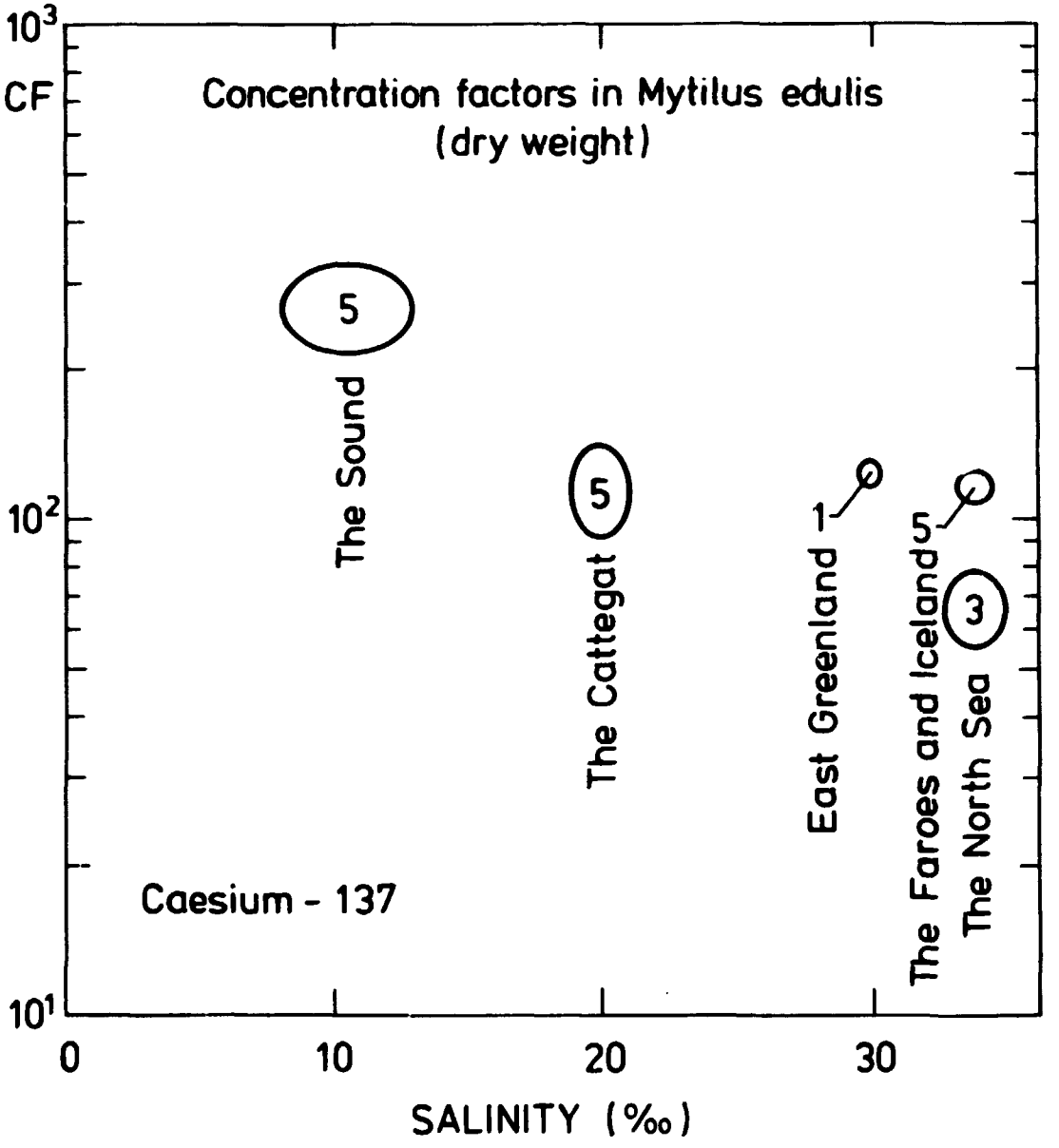

Fig. 3.2.1.9. Concentration factor Mytilus/sea water for ^{137}Cs in the Northern North Atlantic (number of determinations and 1 SE indicated).

In Figs. 3.2.1.8 and 3.2.1.9 the concentration factors calculated for Mytilus edulis are shown for comparison.
(NKA references: 1.2, 1.4, 3.1, 3.2, 3.3, 3.5, 3.6, 3.8, 3.10, 3.11, 3.13, 3.14, 3.15, 3.16, 3.17, 3.18, 3.19, 4.1, 4.2, 4.3, 4.4, 4.5, 4.6, 4.7, 4.8, 4.9, 4.10, 4.11, 4.12, 4.13, 4.14, 4.15, 5.1, 5.2, 5.4, 5.5, 5.7, 5.9, 5.12, 5.14, 5.15, 5.16, 5.17, 5.18, 5.19, 5.29, 5.27.)

3.2.2. Baltic studies with F/S Gauss

In May-June 1983 and in Dec. 1984 professor Hans Kautsky from the German Hydrographic Institute in Hamburg invited Nordic colleagues on two Baltic cruises with F/S Gauss. The participating Nordic laboratories were the Finnish Centre for Radiation and Nuclear Safety, Helsinki, Finland, Lund Unviersity, Sweden, AB Energitechnique, Studsvik, Sweden and Risø National Laboratory, Roskilde, Denmark.

The purpose was to get an immediate view of the radioactive pollution of the Baltic Sea and to intercalibrate sample equipment and procedures.

The analysis on the samples has not yet been completed, but some preliminary results have already been reported.

The study comprised measurements on benthos from various parts of the Baltic area. The levels in Macoma baltica varied between 0.5 and 3.0 Bq ^{137}Cs kg^{-1} dry weight and crustaceans between 4.1 and 15.0. The corresponding Pu concentrations were 0.02-0.06 and 0.03-0.38 Bq $^{239,240}Pu$ kg^{-1}, respectively. In general, the benthos showed higher ^{137}Cs concentrations at the low salinity stations than at those with higher salinity. The benthic communities contain only a minor part of the radionuclides present in the sediments.

A number of water samples were analysed for ^{99}Tc. The Baltic surface water contained 68 $\pm$ 11 mBq m^{-3} and the bottom water 95 $\pm$ 11 mBq m^{-3}. From this the concentration factor for Fucus was found to 25000 or four times less than found in the Cattegat.

Large seawater samples (~ 2 m^3) collected in the Danish Straits in May 1983 contained 0.12 Bq ^{60}Co m^{-3}. From this and the levels in neighboring Fucus samples the concentration factor in the Cattegat was estimated to (1-2) $\times$ 10^4.

Seawater samples collected under the cruise in 1983 were used for a Baltic intercomparison exercise run by the IAEA, Monaco laboratory.
(NKA references: 3.2, 3.14, 3.16, 4.10, 5.13, 5.18, 5.21, 5.27.)

3.3. Field experiments

3.3.1. Calibration of recipient

These studies have been carried out at Finnish as well as at Swedish nuclear power plants. Samples of fucoids (and other algae), mytilus, sediments and seawater have been collected.

3.3.1.1. Bioindicator studies at Finnish power plants. Between 1 and 10 km from the discharge point the ^{60}Co activity at Loviisa was proportional to $X^{-1.8}$, where X was the distance in km from the discharge point. At Olkiluoto the ^{60}Co in Fucus followed $X^{-0.8}$.

The transfer factor from ^{60}Co discharge to Fucus at 1.4 km distance was 3 Bq per GBq a^{-1} at Olkiluoto and 30 Bq kg^{-1} per GBq a^{-1} at Loviisa. These values may be compared with 50 for Barsebäck and 1.2 for Ringhals. Although these calculations are based upon relatively few measurements the Finnish values are compatible with those observed at Swedish nuclear plants. (NKA references: 3.18, 3.19, 4.12, 4.13, 5.7, 5.10, 5.11, 5.12, 5.20.)

3.3.1.2. Speciation studies at Swedish power plants. As the transfer factors to Fucus (Bq kg^{-1} (GBq $a^{-1})^{-1}$) show significant variations between the various Swedish nuclear power stations, (cf. Table 3.3.1.2.1.), the importance of the chemical form of the liquid waste for the 3 power stations: Ringhals, Barsebäck and Oskarshamn has been investigated. Effluents from the 3 stations were diluted with Cattegat seawater in 3 large tanks. Fucus was grown in the tanks and the water was examined. The experiment gave no clear answer to the differences observed in the field. However, it is believed that it plays a role that some of the activity discharged from Ringhals is present in form of sedimentable material, and chemical impurities in the seawater at the various power plants may also have an effect on the transfer factors to Fucus.
(NKA references: 5.8.)

3.3.1.3. Bioindicator studies at Swedish power plants. Since 1977 Fucus samples have been collected 2-3 times every year from one location at Barsebäck and two at Ringhals. The concentrations of various radionuclides in the Fucus have been related to the discharges in the preceeding 12 months, and transfer factors have been calculated. The mean values are shown in Table 3.3.1.3.1.

Table 3.3.1.3.1. Transfer factors (without a decay correction) to Fucus vesiculosus collected 1.4 km from the outlet: (Unit: Bq kg^{-1} d.w. (GBq y^{-1})$^{-1}$

Isotope	Barsebäck	Ringhals*
^{60}Co	50±7 (n=19)	1.2±0.3 (n=20)
^{58}Co	22±4 (n=18)	0.8±0.3 (n=20)
^{54}Mn	33±4 (n=19)	1.2±0.1 (n=20)
^{65}Zn	46±7 (n=19)	5.5±2.1 (n=20)
$^{110}Ag^{m}$	11±4 (n=11)	7.3±1.0 (n=19)
^{51}Cr	4±1 4)	0.1 (n=1)

*Mean of a station at 1.1 and one at 1.9 km from the outlet. The error term is 1 S.E.

The transfer factors predict the levels in Fucus if the discharges are known, but may also be used to estimate the discharges if only the concentrations in Fucus are known. A more refined model has recently been made (cf. 3.5.3.).

Concentration factors for Fucus vesiculosus and Mytilus edulis (soft parts) from Ringhals have been calculated on the basis of mean transfer factors from discharge to seaweed (cf. Table

3.3.1.3.1.) and on measured ratios between Fucus and Mytilus as shown in Table 3.3.1.3.2.

Table 3.3.1.3.2. Concentration factors at Ringhals (salinity 19‰)

	^{60}Co	^{58}Co	^{54}Mn	^{65}Zn	$^{110}Ag^{m}$	^{51}Cr
Fucus vesiculosus						
dry weight	4×10^{4}	2.5×10^{4}	5×10^{4}	15×10^{4}	30×10^{4}	0.4×10^{4}
fresh weight	9.5×10^{3}	5.5×10^{3}	11.5×10^{3}	35×10^{3}	65×10^{3}	1×10^{3}
Mytilus edulis						
dry weight	1.5×10^{4}	0.65×10^{4}	0.8×10^{4}	18×10^{4}	20×10^{4}	-
fresh weight	2×10^{3}	0.9×10^{3}	1.1×10^{3}	25×10^{3}	30×10^{3}	-

At Barsebäck the activity (^{60}Co) in fucoids decreases northward with distance after the relation $X^{-1.4\pm0.1}$ and at Ringhals the decrease in all directions follows $X^{-0.89\pm0.09}$ (X is the distance in km from the outlet). The Ringhals values are calculated within 5 km from the discharge point and those from Barsebäck within 125 km.
(NKA references: 1.4, 1.9, 2.1, 2.3, 3.5, 3.7, 3.17, 4.1, 4.4, 4.7, 4.10, 4.14, 4.15, 5.2, 5.3, 5.6, 5.9, 5.14, 5.22, 5.25, 5.28.)

3.3.1.4. Growth and uptake studies with fucoids. Finnish studies have shown that plutonium and ^{60}Co, ^{65}Zn and ^{54}Mn showed higher concentrations in the lower parts of the Fucus plants than in the growing tips. Cesium-137 was homogenously distributed. The effect of washing or brushing was modest.

Swedish studies at Ringhals showed that the concentrations of ^{137}Cs, ^{65}Zn and ^{60}Co did not differ with the age of the Fucus plants (1-2-3 years). It is concluded that Fucus shows a rapid response on waterborne radioactive pollution.

Marine botanical studies at Barsebäck shows (in accordance with the Finnish results) lower concentrations of ^{60}Co, ^{54}Mn and ^{65}Zn in the growing tips of Fucus than in the lower parts of the plant, but in the case of $^{110}Ag^{m}$ the opposite effect occurs. At a given location the coefficient of variation C.V. (relative S.D.) for ^{60}Co in Fucus was 20%, for ^{137}Cs: 30% and for ^{40}K: 21% (24 samples). If the $^{60}Co/^{40}K$ and $^{137}Cs/^{40}K$ ratios were considered the C.V. becomes 11% and 21%, respectively.
(NKA references: 3.18, 3.19, 4.12, 4.13, 4.14, 4.15, 5.4, 5.7, 5.9, 5.12, 5.20.)

3.3.2. The biotestfacility at Forsmark, Sweden.

Various experiments with Fucus and Mytilus have been initiated in the biotestlake in a co-operation between SNV and RISØ (see 3.4.2.).

3.4. Laboratory experiments

These studies belong to the so-called REK5A project, but they were initiated under the bioindicator project and shall therefore shortly be mentioned here.

3.4.1. Fucus studies

Initial rates of accumulation in Fucus gave the following mean concentration ratios (Bq g^{-1} dry/Bq ml^{-1}) after 24 hours: Co: 1000; Zn: 1800; Cs: 70; Ce,Eu,Am,Cm: 2500; Np: 40; Pu(IV): 700. In a single experiment Pu(VI) showed a value of 250 and Tc: 4800. There is a pronounced variation with time. Thus, the 9 experiments showed variations (S.D.) of 30-60% for Pu(IV), Am, Np, Eu, Ce, Zn and Co, whereas the variation for Cs was only 15%.

The salinity effect on the accumulation after 24 hours (15 o/oo/ 30 o/oo) was a factor 2.5 ($\pm$ 30%, S.D.) for Cs, Ag, Zn, Co and

Mn. The temperature effect (20°C/3.5°C) was 2.5 (± 30%, S.D.) for Cs, Zn and Co and a light effect (light/dark) of 4.4 (± 15%, S.D.) was recorded for Cs. There was no significant salinity effect for Ce, Eu, Am, Cm, Fe and Co; no temperature effect for Ce, Eu, Am and Cm and no light effects except as mentioned for Cs and in some experiments Zn and Co.

At two instances (winter 82-83 and spring-summer 84) long-term loss experiments were run with Fucus vesiculosus in Roskilde Fjord. After an initial loss, Zn, Co, Ag and Tc were lost very slowly ($t_{1/2}$ > 0.5 year). For Cs a biological half-life of 35 days was recorded during spring, whereas it was twice as long during the winter. The lanthanides Ce and Eu were lost with a half-life of ~ 20d. All these biological half-lives were measured excluding growth dilution. During the spring experiment the biomass doubling time was measured as two months corresponding to a two-month half-life of activity concentration in the tissues. Loss of the α-emitters was studied in sections of plants sampled during the experiments. Results suggest that Cm, Pu(IV) and especially Pu(VI) are retained better than Ce and Eu. Thus, the two lanthanides can probably not be used as actinide-analogues in Fucus.
(NKA references: 2.2, 3.7, 5.2, 5.6, 5.25.)

3.4.2. Mytilus studies

A one-year excretion study was run in the Bothnian sea (5 o/oo salinity) at the Forsmark nuclear power plant and "biotest" area, Sweden, from October 1983. A distinct seasonal effect on loss-rates was found. After the initial loss, most nuclides (Eu, Ce, Tc, Zn, Co, Mn) were apparently not excreted at all during the long, cold winter, whereas biological half-lives of 30-60 days were recorded for Mytilus soft parts during the next summer. Results for the α-emitters Pu(IV), Am and Np will appear later. In an accumulation experiment it has been seen

that Am and Cm behave exactly similar in seawater and mussels. Great variation between individual animals was identical for the two nuclides. Cm can therefore be used as an Am-analogue when found feasible.
(NKA references: 2.2.)

3.5. Models

3.5.1. A local dispersion model for Barsebäck

A current measuring programme near Barsebäck was set up, and analysis of the results showed that if periods longer than 7 hours were eliminated no significant correlation could be found between locations 900 meters apart. This is used in the simulation model, which also is based on autoregressive models estimated from the current velocity data. The model gives a description of the dilution in the area north of Barsebäck. The results have been verified by bioindicator measurements, i.e. the dilution follows the expression $X^{-1.4\pm0.1}$ (cf. 3.3.1.3. & 3.5.2.)

In the Appendix is given a more detailed description of the dispersion model for the Barsebäck area.
(NKA references: 2.1.)

3.5.2. The cobalt-60 fucus model

This model is based upon ^{60}Co measurements of Fucus vesiculosus collected in the Danish Straits in 1982 and 1983 (cf. Fig. 3.5.2.). The concentration, C, of ^{60}Co in Fucus, at a distance X km from Barsebäck is described by

$$C\ (Bq\ kg^{-1}\ dry\ weight) = 1200\ (\pm 400)\ X^{-1.4(\pm 0.1)}$$

If we assume a concentration factor from seawater to Fucus for ^{60}Co of 2×10^4, a mean depth in Cattegat above the halocline of 12 m, and a fraction 1/10 to indicate the area represented by

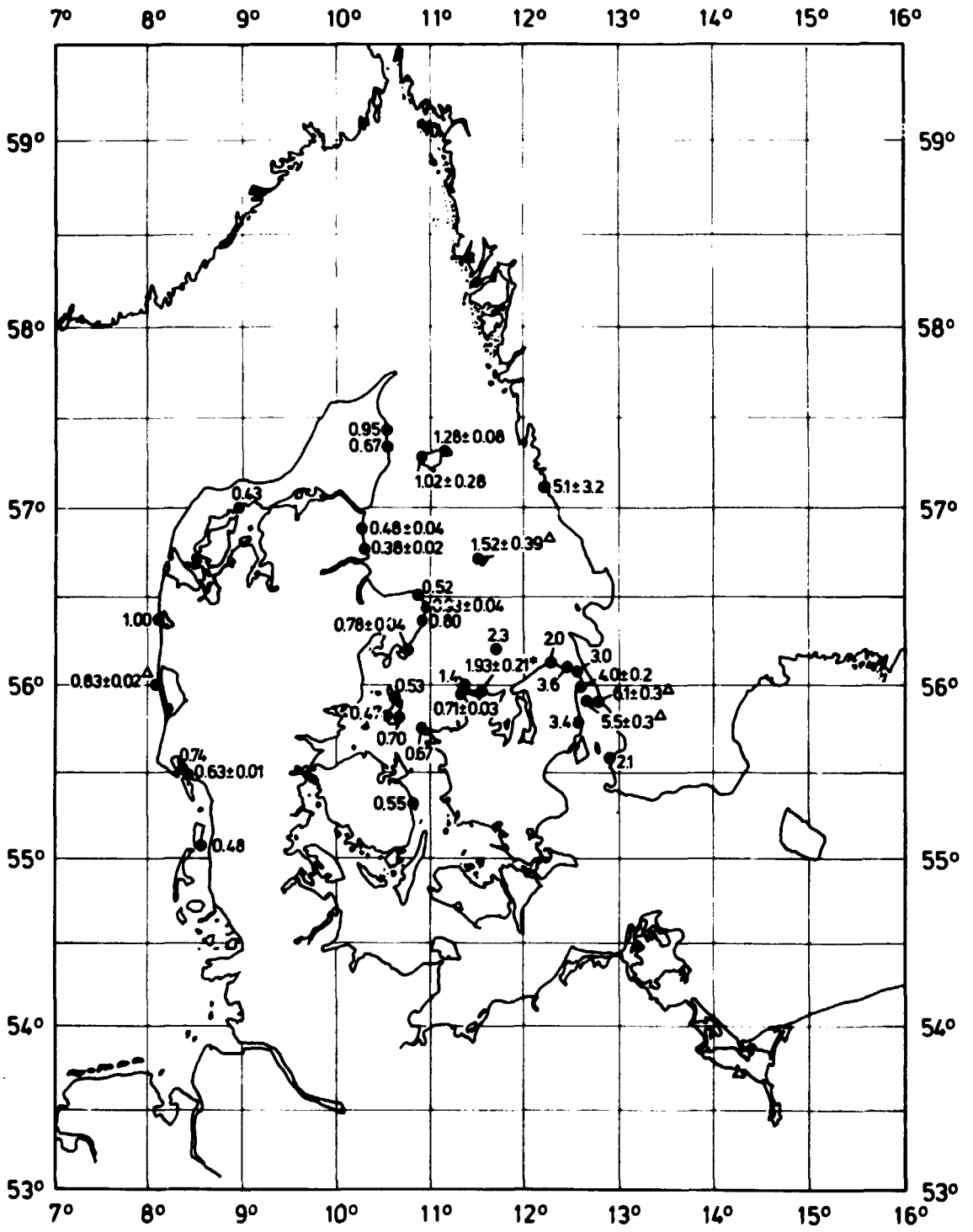

Fig. 3.5.2. Cobalt-60 in Fucus in the Danish Straits in 1983 (Bq kg^{-1} d.w. ± 1 SE).

Cattegat compared with a circle of radius 270 km (the distance from Barsebäck to Skagen), the inventory of ^{60}Co in the Cattegat, I is then estimated as:

$$I = \int_1^{270} 2\pi R \times (2 \cdot 10^4)^{-1} \times 1200\ R^{-1.4} \times 0.012 \times 0.1 \times 10^{12}\ dR$$

$$= 21 \times 10^9 \text{ Bq } ^{60}Co$$

If this inventory is compared with the annual mean discharge of ^{60}Co from Barsebäck, i.e. 108×10^9 Bq, a mean residence time in Cattegat of $\frac{21}{108}$ year = 0.2 year is calculated this is in agreement with the estimate based upon waterflow (Nielsen G.B. et al.: The Belt Project, The National Agency of Environmental Protection, Denmark (1981)).

From this model it is furthermore possible to calculate the annual dose in 1982-83 from ^{60}Co from Barsebäck to the critical individual in the Cattegat region who consumes 100 kg fish per year from the Cattegat. The dose becomes 7×10^{-9}Sv or 0.2% of the dose from ^{137}Cs (from Sellafield discharges) or 7×10^{-6} times the background radiation.
(NKA references: 1.4, 1.9, 2.1, 2.2, 2.3, 3.5, 3.17, 4.1, 4.4, 4.7, 4.10, 5.14, 5.22.)

3.5.3. The SENSI model

This model describes the uptake of radionuclides in Fucus vesiculosus throughout the year. The model is based upon laboratory experiments as well as field studies. The overall idea is to construct a "sensitivity matrix" showing month-by-month a relative factor to be multiplied with water concentrations or discharge data in order to give a relative number for the concentration expected in the bioindicator from discharges each month preceeding the sampling. Figure 3.5.3. shows how predicted values compare with those observed. This model will be further improved,

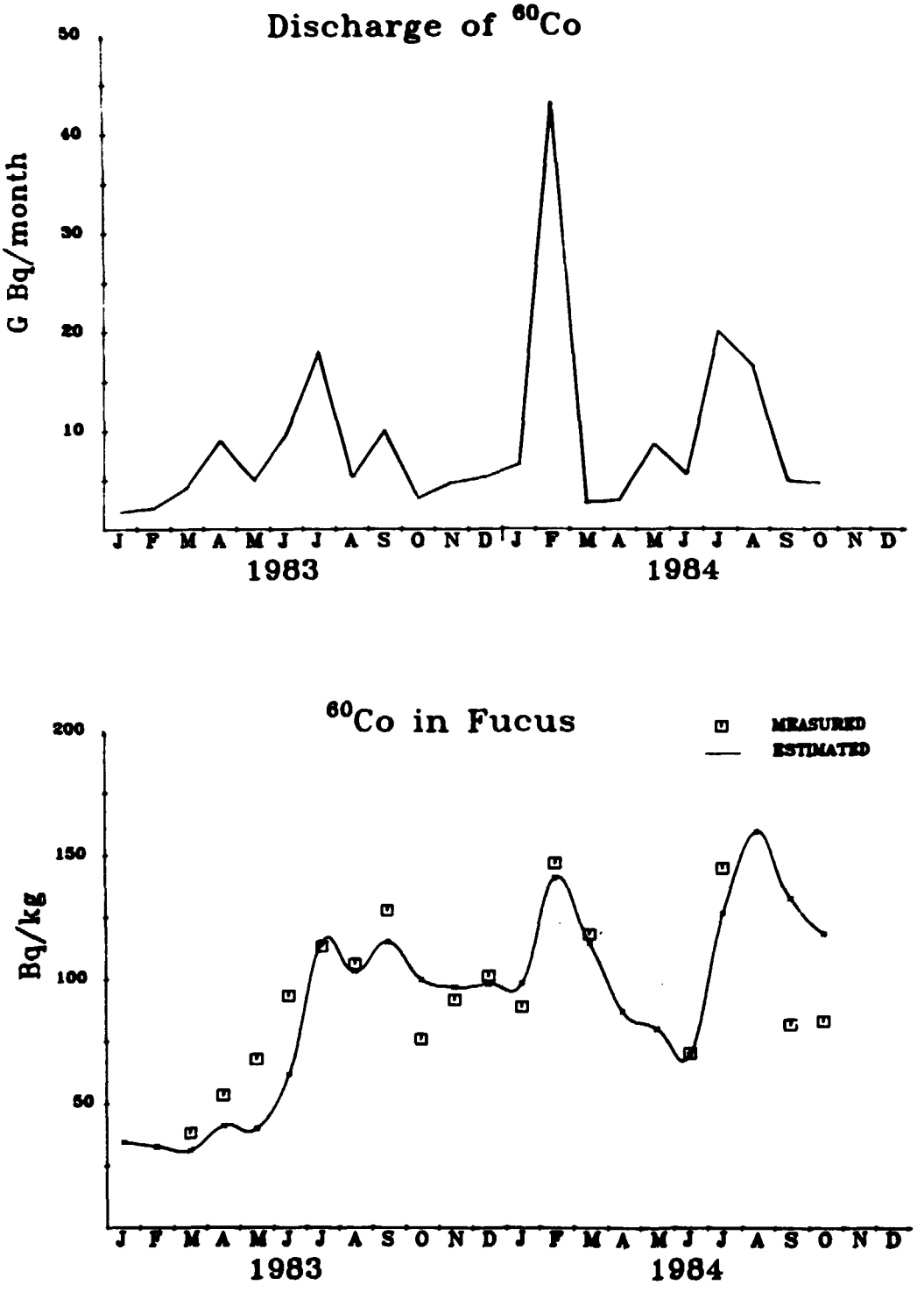

Fig. 3.5.3. Measured versus predicted ^{60}Co levels in Fucus according to the SENSI model.

when more data for growth during every month of the year has been collected.

(NKA references: 5.25.)

3.5.4. Baltic compartmental model

NRPB (Clark, M.J. et al.: NRPB-R-109, National Radiation Protection Board, 1980) has made a model to calculate the exposure from radioactive discharges into the coastal waters of Northern Europe. In this model the Baltic Sea, the Danish Straits and the Skagerak are considered together as one compartment only. Evans (S. Journ. of Environmental Radioactivity 2,41 1985) has improved this model for the Baltic area by adding a number of new compartments. Evans's work has not been a part of the present NKA project, but it is closely related to the interpretation of the data collected during this project and also to the models mentioned above.

Figure 3.5.4.1. and 3.5.4.2. show the time variation in the water concentrations of a conservative element (^{99}Tc) and of ^{60}Co respectively after a momentary release to the Sound.

We have also made a simple calculation for a release from Barsebäck. We assume two compartments: the North (7×10^4 km^3) and Baltic Seas (2×10^4 km^3) the mean residence times of water are 3.5 and 25 years respectively. In the Sound the northgoing water is two times that of the south running. Hence the collective doses from fish consumption can be calculated if we know the release, the catch of fish and the concentration factors between water and fish (Gjørup et al. Risø-R-462, p. 169-173, 1982).

(NKA references: 3.4.)

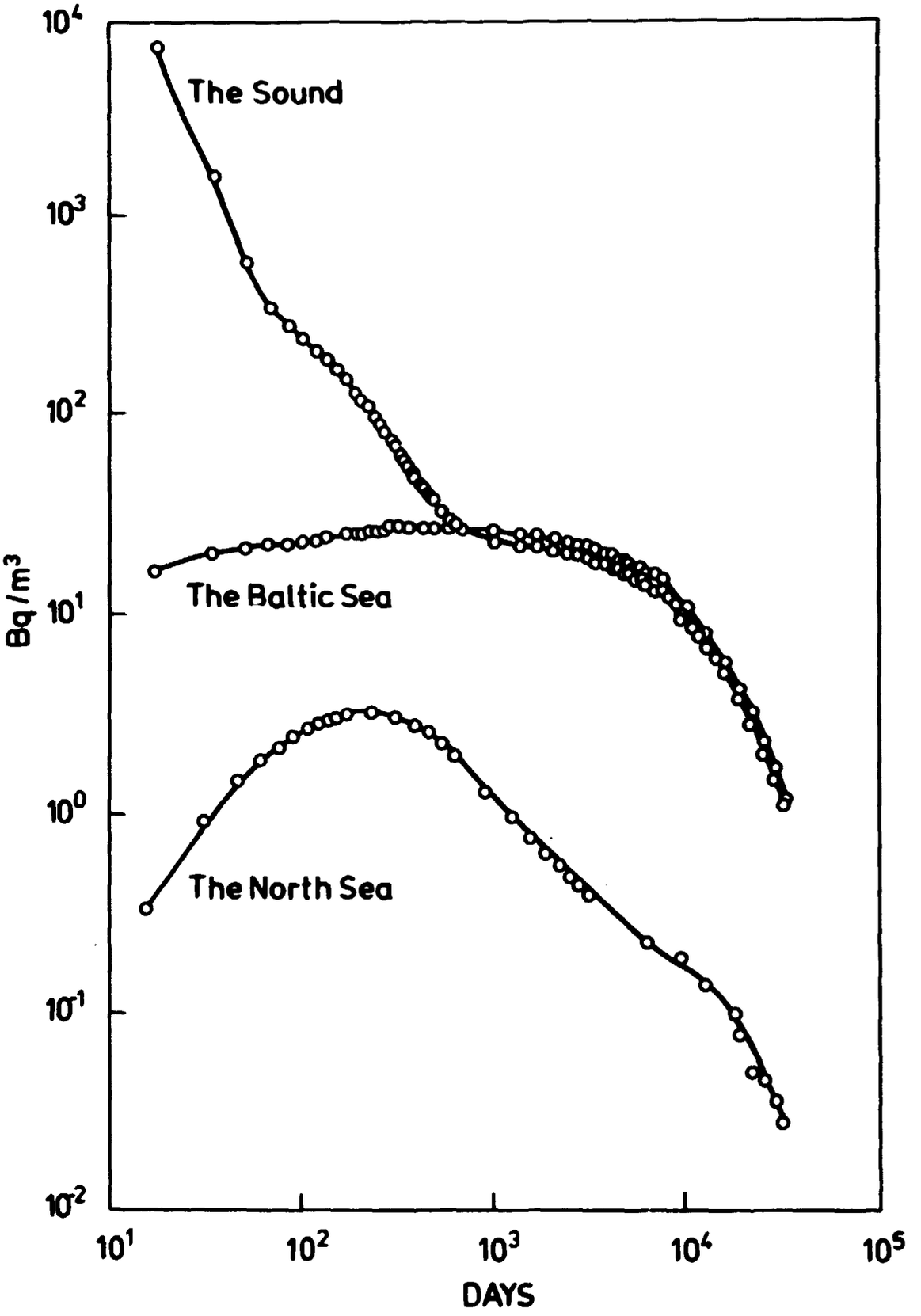

Fig. 3.5.4.1. Water concentrations of ^{99}Tc according to the compartment model after a release of 1 PBq to the Sound.

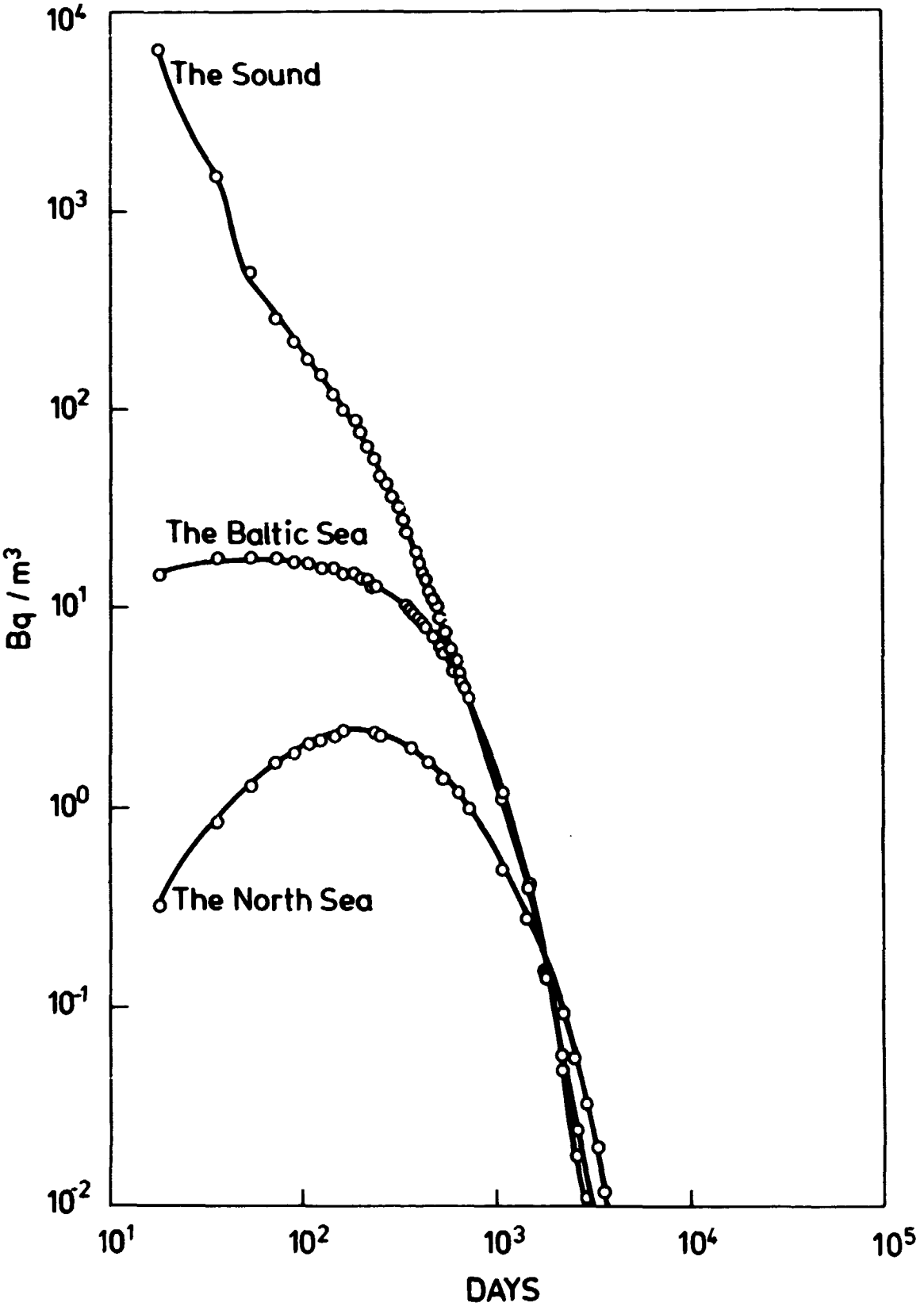

Fig. 3.5.4.2. Water concentrations of ^{60}Co according to the compartment model after a release of 1 PBq to the Sound.

3.5.5. Time dependent Fucus model

The radionuclide concentration C(t) in Fucus can be expressed as:

$$\int \frac{dC(t)}{dt} = a \cdot A(t) - (K+\lambda)C(t)$$

where a is an accumulation factor of a nuclide, describing the increase of activity concentration in Fucus compared with seawater.

A(t) is the concentration of a nuclide in seawater surrounding the Fucus plant,

K is the elimination rate of activity of a nuclide,

λ is the disintegration constant of a nuclide.

In Finnish studies a has been found to 10^4 for ^{60}Co and the biological half-life to 75 days.

In the above equation A(t) may, however, also be the discharge rate of a nuclide. If so, a describes the increase in concentration of a nuclide in Fucus after a release of unit activity or in other words the transfer factor from discharge to the plant (Bq kg^{-1}/Bq) = kg^{-1}. Swedish studies at Barsebäck has determined this transfer factor to 2.1×10^{-8} kg^{-1} and the biological half-life $\frac{\ell n 2}{K}$ to 56 days. At Oscarsham and Ringhals, SNV found biological half-lives for ^{60}Co of 44 days and 52 days, respectively.

(NKA references: 3.17, 3.18, 5.3, 5.28.)

4. CONCLUSIONS

4.1. Bioindicator systems

During the project we have identified two important bioindicators, well suited as monitors for radionuclides in the marine environment: The Blue mussel: Mytilus edulis and the Bladderwrack: Fucus vesiculosus. Both indicators are available from nearly all parts of the northern North Atlantic region, and

for many of the radionuclides studied, they show accumulation factors from the water in the order of 10^3 or even higher.

Other brown algae may supplement the Fucus vesiculosus as a bioindicator, e.g. Ascophyllum nodosum, Fucus serratus, Fucus spiralis, Fucus disticus and Laminaria sp.

4.2. Description of the contamination pattern

The radioactive contamination (C) from a source, e.g. a nuclear power plant, decreases with the distance (A) from the source after a power function:

$$C = K \cdot A^{-\beta}$$

where β typically for various locations varies between 0.5 and 2.

This relation with distance may be valid out to distances of several hundred kilometers.

We have in particular followed the distribution of the discharges from Sellafield in the U.K. and have been able to trace this radioactivity over distances of several thousands of kilometers. We have calculated dilution factors between the North Sea and other parts of the Northern North Atlantic and have determined the transit times for water masses.

4.3. Transfer factors

For discharges from the nuclear power plants in Sweden and Finland we have calculated the transfer factors of various radionuclides to Fucus vesiculosus growing at a distance of approx. 1.5 km from the outlet. (Unit: Bq kg^{-1} dry weight per GBq a^{-1}). We have furthermore for the northern North Atlantic calculated transfer factors for ^{90}Sr, ^{99}Tc and radiocesium to the surface water from discharges from Sellafield (Unit: Bq m^{-3} per PBq a^{-1}).

The transfer factors make it possible to predict the contamination pattern in the environment if the discharges are known.

Furthermore, if the discharges are unknown, these may be estimated from measurements of the bioindicators when the transfer factors to these are known. This principle has been used to estimate the unknown discharges of ^{99}Tc from Sellafield prior to 1978.

The transfer factors from Sellafield discharges to the water in the Danish straits have also been estimated. A little less than 1% of the total discharge from Sellafield enters the Baltic Sea. The transit time from Sellafield to the Danish straits is 4 years.

4.4. Doses to man

From our bioindicator studies we have been able to estimate the mean residence time of water in the Danish straits (The Cattegat) to 0.2 years. We may use this information for a calculation of the doses to man from consumption of fish contaminated with discharges from the two Swedish power plants: Ringhals and Barsebäck.

Let us assume that the annual discharge rate to the Danish Straits of radionuclide (n) is (A_n) GBq a^{-1}. If we neglect radioactive decay, the inventory (I) in the Danish straits becomes $0.2\ A_n$ GBq. The volume of water concerned is estimated to $2.75 \times 10^{11}\ m^3$, and the concentration factor from water to fish is (CF_n)Bq t^{-1} (Bq m^{-3})$^{-1}$. Hence the concentration in fish becomes:

$$\frac{0.2\ A_n \times CF_n}{2.75 \times 10^{11}} \quad GBq\ t^{-1}$$

If we assume that a member of the critical group consumes 0.1 t fish per year from the Danish straits and that the annual limit

of intake (ALI_n)GBq yields an effective dose equivalent of 0.05 Sv, the dose to the critical individual becomes:

$$\frac{0.2\ A_n \times CF_n \times 0.1 \times 0.05}{2.75 \times 10^{11} \times (ALI_n)}\ Sv = \frac{A_n \times CF_n}{ALI_n} \times 3.64 \cdot 10^{-15}\ Sv$$

Table 4.4. Annual mean doses from waterborne discharges from Barsebäck and Ringhals to a critical individual consuming 100 kg fish annually caught in the Cattegat (1976-1983).

Nuclide	CF_n fish/water	ALI_n	Annual discharges		Effective dose equivalent	
			Barsebäck	Ringhals	Barsebäck	Ringhals
	m^3/t	GBq	GBq	GEq	nSv	nSv
^{54}Mn	4×10^2	7×10^{-2}	2.46	0.03	0.05	0.19
^{60}Co	10^3	2×10^{-2}	40.0	68.3	7.3	12.4
^{65}Zn	10^3	10^{-2}	5.97	42.9	2.2	25.6
^{134}Cs	10^2	3×10^{-3}	10.2	18.9	1.2	2.3
^{137}Cs	10^2	4×10^{-3}	14.8	27.1	1.3	2.5
Sum					12.1	33.0

The annual mean productions of electricity from Barsebäck and Ringhals were by 1983 0.8 $GW_e a$ and 1.2 $GW_e a$, respectively. Hence the mean critical indivual dose via fish consumption from Barsebäck became 15 nSv per $GW_e a$ and from Ringhals 27 nSv per $GW_e a$.

In the Cattegat the annual total fish catch is 0.12 Mtons. Of this approx 36% are fish for consumption, i.e. 0.043 Mtons, and of this only half actually enters the human diet, i.e. 0.02 Mtons. Hence the annual collective mean doses from consumption of fish caught in the Cattegat becomes 0.0024 manSv from Barsebäck and 0.0065 manSv from Ringhals or in total ~ 0.01 manSv. This figure may be compared with the annual doses received in the later years (1983) from ^{137}Cs discharged from Sellafield.

The concentration in fish in the Danish waters of ^{137}Cs derived from Sellafield is 3.6 Bq kg^{-1} i.e. the intake with 0.02 Mtons fish becomes 0.072 GBq ^{137}Cs corresponding to a collective effective dose equivalent of 0.9 manSv or two orders of magnitude higher than that received from Barsebäck and Ringhals together.

The collective dose from naturally occurring ^{210}Po in the 0.02 Mtons fish, which contain 1 Bq ^{210}Po kg^{-1}, is 10 manSv, and global fallout in this fish catch yield approximately 0.1 manSv of 10 times more than the dose from Ringhals and Barsebäck.

4.5. Influence of environmental factors

It has been shown that the accumulation of most radionuclides by Fucus vesiculosus is higher in low-salinity waters than in ocean water. The initial accumulation of Cs, Zn and Co is higher at high temperatures than at low; there is a higher initial accumulation of Cs in light than in darkness. The biological half-life of most radionuclides is higher during the winter than in summer. This is the case for Fucus as well as for Mytilus.

4.6. Models

Local and semiregional models have been set up in the Danish Straits around Barsebäck and Ringhals. The models describe the dispersion of radionuclides discharged to the water, and show e.g. how ^{60}Co in Fucus varies with distance from the discharge point. The so-called SENSI model describes the seasonal variation in the uptake of radionuclides in Fucus. The Baltic com-

partment model has been used for a description of the contamination of the Baltic Sea with Sellafield effluents.

In the future modelwork it is essential that the models set up are well established on a basis of actual measurements in the environment or in the laboratory, otherwise such models will be of only theoretical interest and should not be used for practical purposes.

5. FUTURE STUDIES

5.1. Improvement of database

The calculation of doses based upon bioindicator studies is encumbered with a number of uncertainties, which have been identified during the project. Future work will focus on these uncertainties in order to minimize them.

The transfer factors between discharges and concentrations found in bioindicators may be more reliably determined if the seasonal variations in the uptake are included, as was recently done in the SENSI model.

We need more reliable determinations of concentration factors especially in fish in low salinity waters. Such studies may be carried out in the biotest facility at Forsmark. It would however be desirable if similar experiments could be carried out in the higher salinity regime found at Ringhals.

The exponent (β) determined in the power function $C = K \cdot A^{-\beta}$, which describes the decrease in activity concentrations with distance, may vary with time. This should be investigated by occasional remeasurements in the coming years. A special aspect

has to be considered in the Danish Straits. The power function for the water concentrations may be different from the one observed for the activity concentrations found in Fucus, because the concentration factor between Fucus and water usually increases with decreasing salinity. If we e.g. consider Barsebäck, for which we have determined β =1.4 from Fucus measurements, we may expect a lower value for seawater, because the salinity increases, when we move northward with the current from the Sound to the Cattegat.

During our studies of ^{60}Co in the Nordic waters we have found indications of sources other than those arising from Swedish and Finnish nuclear power plants. These partly unidentified sources should be known in order to improve our models and dose calculations.

5.2. "Exotic" radionuclides

The present bioindicator project has focussed on those radionuclides which are relatively easy to measure. However, a number of radionuclides present in the environment are difficult to determine because their radiochemical identification is problematic. This can e.g. be because they emit only low-energy β-radiation or because no suited yield tracers are available. Ni-63 is a typical example of such a radionuclide. But we may also mention long-lived radionuclides such as ^{53}Mn, ^{59}Ni, ^{79}Se, ^{93}Zr, ^{92}Nb, ^{94}Nb, ^{93}Mo, ^{99}Tc, ^{126}Sn, ^{129}I and ^{237}Np. The aim of further studies will be to develop counting equipment and analytical methods for determining and measuring some of these radionuclides in low concentrations in environmental samples.

5.3. The Biotest facility at Forsmark

Only in the very last part of the project the biotest facility has been available for the bioindicator studies. This facility is however so unique that it will be highly desirable that future bioindicator studies in the Nordic countries can benefit from the biotest lake. An important aspect of using this facility is the low salinity. This means that the accumulation of radionuclides in many organisms will tend to yield maximum values. Hence we may get an upper limit for concentration factors, when we carry out experiments in the biotest lake.

5.4. Terrestrial bioindicators

In this project we have focussed on the marine environment. It would however be desirable also to include the terrestrial environment in order to obtain a more reliable evaluation of the doses from terrestrial food chains contaminated with airborne effluents from nuclear facilities. We know that moss and lichens are excellent bioindicators for airborne pollution, but there may be other terrestrial bioindicators, e.g. fungi and ferns. This may be a part of a future bioindicator study.

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7. APPENDIX - A Dispersion Model -

by Søren Boelskifte

7.1. Introduction

Transport and dispersion models for the marine environment are used in estimating doses from the aquatic food chain. The initial purpose of the present investigation was to evaluate some of the models available with emphasis on their need for reliable input data, their complexity, and the time and length scale they cover. Another purpose relates to the construction of a local dispersion model for the area near Barsebäck; it was to get a picture of the correlation structure of the current in this area (see below). Also a statistical description of the current is needed in connection with the dispersion model. This description is based on time series analysis (Box-Jenkins method, see below), and the study is made mainly to test this method on the actual problem.

7.2. Dispersion models

The most important question to be answered before a model is constructed is what time- and length scales it has to cover. Is it the behaviour of the discharge during the initial mixing that is important, or is it the long time dilution over, e.g., thousands of kilometers? Each length scale needs its own model as the physical processes involved are completely different. Also the purpose of the model is important, for example: Should we consider continuous releases or those that are momentaneous? Should we emphasize maximum concentrations or the mean?

Appendix

Concerning radioactivity and collective doses the box-model developed at NRPB (Clark et. al, 1980) is often referred to. It covers the "North European Waters" and gives a rough description of the transport in this area. It is not detailed for the Danish straits and the Baltic, but Evans (1984) has improved the model by constructing boxes for different parts of the Baltic and by including two-layer boxes, thus reflecting the strong stratification in the Baltic.

This type of model cannot describe the situation in the Sound in detail, where the exchange rate is high and no data for making a subdivision would be accessible. For an accidental release from Barsebäck the dose depends to a great extent on the current direction at the moment of release, as discussed by Boelskifte (1983).

For releases during a long time period empirical models exist for the Sound and Kattegat based on measurements on bioindicators (Mattsson et. al (1980), Aarkrog et. al (1982), Boelskifte (1985a)), and they all lead to a similar expression for the concentration of radionuclides in Fucus vesiculosus as a function of distance x from Barsebäck

$$c(x) = \alpha\, x^{-\beta}$$

where α depends on the release and isotope and β is estimated at 1.4.

Other types of models are those based on diffusion theory and using the diffusion constant D as an important parameter. Practical examples of this method applied on radionuclides can, for instance, be found in Niemczyk et. al (1981). The general equation is the following:

$$\frac{dc}{dt} = -V_x \frac{dc}{dx} - V_y \frac{dc}{dy} - V_z \frac{dc}{dz} + \frac{d}{dx} D_x \frac{dc}{dx} +$$

$$\frac{d}{dy} D_y \frac{dc}{dy} + \frac{d}{dz} D_z \frac{dc}{dz} - \lambda c + s(t)$$

where λ is a decay constant and s(t) are sources.

The equation is used to describe diffusion in the Great Lakes in USA.

A similar approach has been made by the Danish Hydraulic Institute (DHI, 1977). This model is very complex and needs a lot of input data and computer time and space, but if that is accessible it is a most reliable model.

A combination of a deterministic and a stochastic model is found in Bork (1977). It is applied on the lake Vänern in Sweden where current measurements are made every 10 minutes. In the simulation scheme single particles are looked upon each having a velocity which is the sum of a deterministic and a random, turbulence imitating part

$$\frac{dx}{dt} = U + a\ P_u$$

where U is the mean velocity, a a random number between -1 and +1, and P_u a known turbulence scale. One drawback of this method among others, is that no correlation between particles and between two time steps are considered.

From the foregoing examples it is obvious that much effort would be saved if the simulations could be based only on current measurements, i.e. without tracer experiments.

Appendix

7.3. Correlations and current measurement

Two particles in water close to each other will have very similar movements. The further apart they are the less correlated will be their movements and therefore their separation will increase rapidly. This well-known phenomenon explains the importance of the correlation structure for the current in a given area in a study of the dilution mechanics (turbulence). For an area north to Barsebäck a measuring programme was set up to get a detailed picture of the correlation in this area, which is important for the initial mixing of effluents from the power plant.

A more comprehensive description of the theoretical background and the measurements is found in Boelskifte, (1985b).

Five current meters (ST1, ST2, ST3, ST4 and ST5) were used and relatively placed as shown in the figure

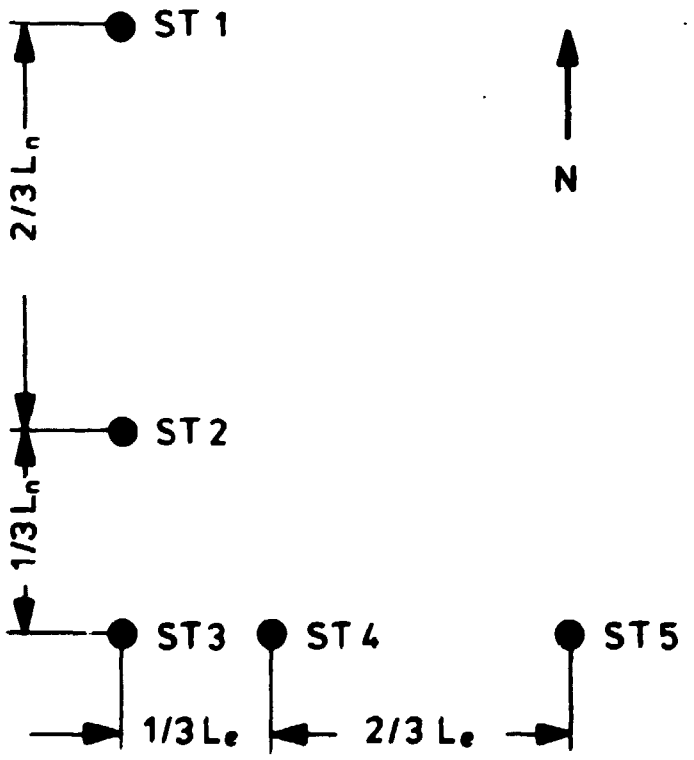

Fig. 7.1. Relative positions of the five current meters.

This configuration was used to optimise the number of different distances covered. The first week L_N was equal to 900 meters and L_E was equal to 300 meters. The second week L_N was 300 meters and L_E was 90 meters, i.e. after the first week all current meters were moved closer together, but still maintaining the configuration shown.

For V, the current velocity in a given direction, the crosscorrelation between the velocity at two points X and Y is defined as

$$\rho_{x,y}(s) = \frac{E\left[(V_x(t) - \bar{V}_x) (V_y(t-s) - \bar{V}_y) \right]}{\sigma_x \quad \sigma_y}$$

where an overbar indicates mean value, σ^2 the variance and both s and t the time. Thus the correlation is a function of time, but the following results all refer to time zero (s = o), where the correlation often has its maximum.

The correlation between two points is interpreted as a longitudinal one for the velocity components following the connection line between the two points and a transverse one (transverse to the connection line). These two components are obtained easily as long as the meters are placed in a north-south or east-west direction.

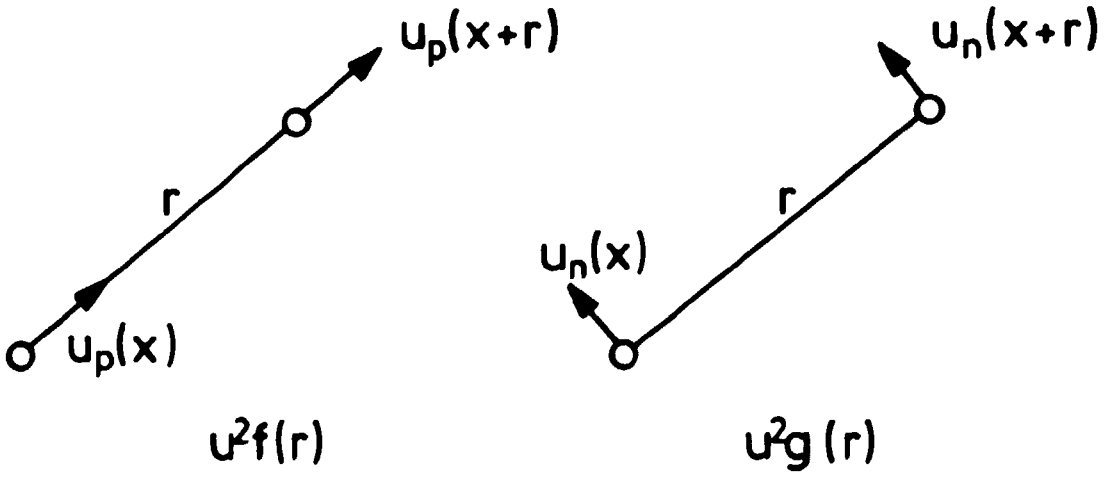

Fig. 7.2. A symbolic representation of the longitudinal and transverse correlations.

Apart from dependence of distance the correlation is a function of the frequency. An example of a power spectrum for the current velocity is shown below.

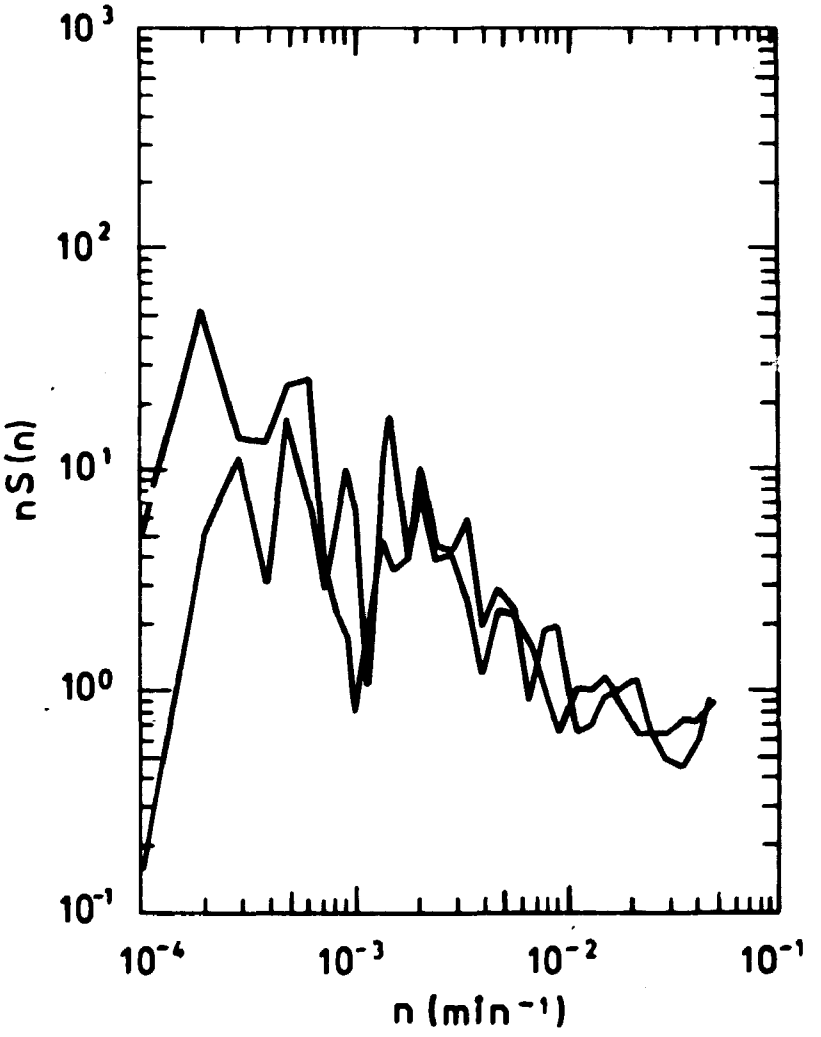

Fig. 7.3. Power spectrum for the current velocity in the north direction.

Most of the energy is contained in the low-frequency part, and this part is not especially relevant to turbulent diffusion, but represents changes in the current with a period of one day or even longer, which is of no significance in relation to diffusion.

An example of the effect of filtration is shown in Fig. 4. Three series are shown together: the lowpass part, the original current, and the turbulent fluctuations.

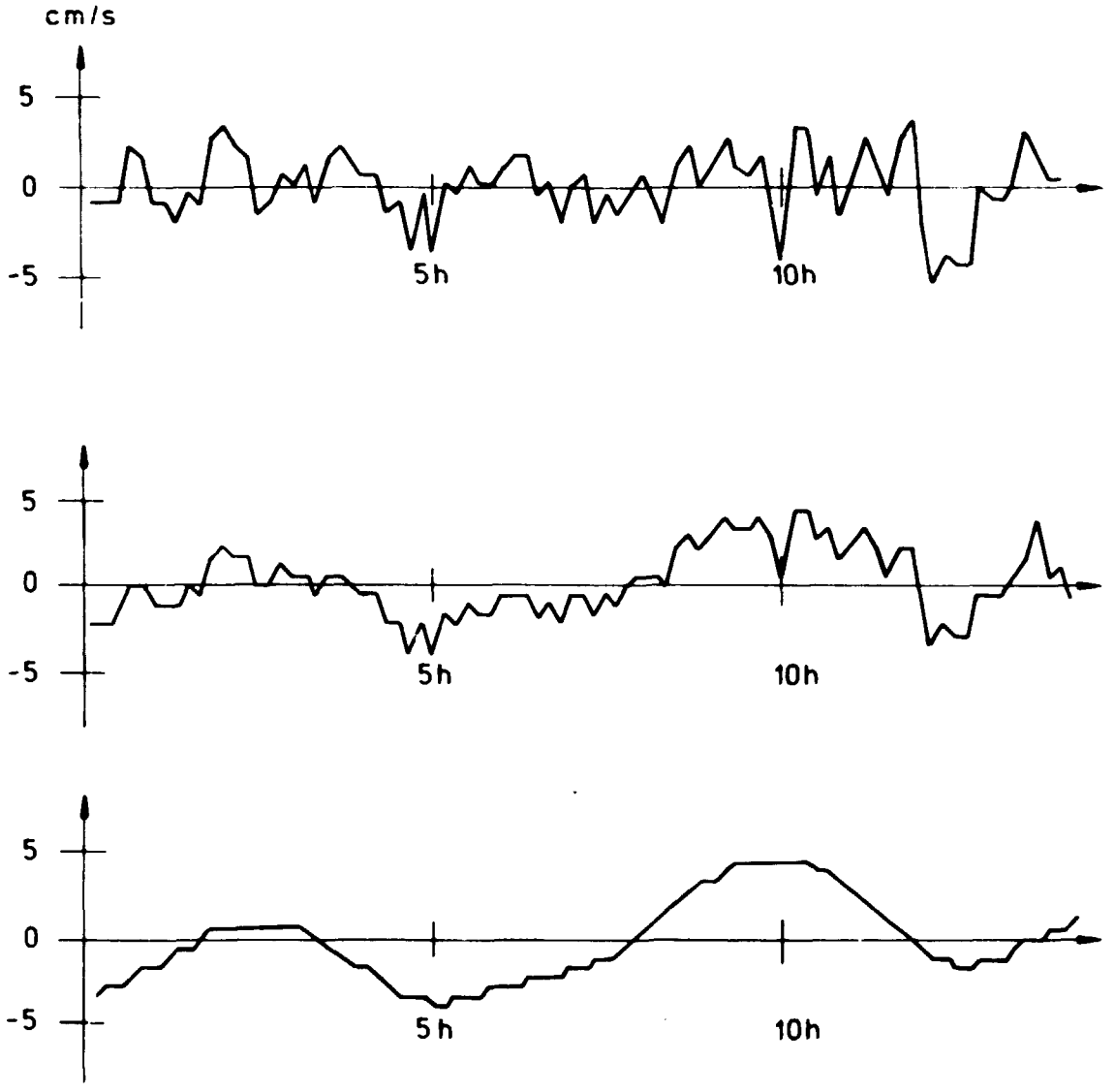

Fig. 7.4. An example of division into mean current and turbulent current.

The highest frequencies in the signal can be separated using a digital high-pass filter, i.e. a signal analyser allowing only the high fr_quencies to pass through. A characteristic property for the filter is the cut-off frequency below which the signal cannot pass. The choice of this frequency will influence the correlations, the higher the cut-off frequency (i.e. the smaller the periods that are observed, or equivalently, the smaller eddies in the turbulence) the smaller the correlation.

Appendix

In Table 1 the correlations on the raw data and after different filtering are shown. Here the dependence between cutoff frequency and correlation is obvious.

Table 1. Correlations, before and after filtration. ST1-ST3 N means the correlation in the north component between Stations 1 and 3

	Cut-off frequency	0	$(33.5h)^{-1}$	$(24h)^{-1}$	$(17h)^{-1}$	$(10h)^{-1}$	$(8.5h)^{-1}$	$(7h)^{-1}$	$(3.5h)^{-1}$	$(1.8h)^{-1}$	$(0.5h)^{-1}$
Combinations											
	ST1-ST3 N	.79	.42	.31	.23	.16	.16	.18	.19	.12	.08
	ST1-ST2 N	.73	.47	.36	.26	.14	.12	.10	.09	.13	.13
	ST2-ST3 N	.76	.55	.45	.39	.31	.28	.23	.10	-.01	-.20
	ST1-ST3 E	.24	-.01	-.01	-.05	-.10	-.10	-.09	-.06	-.07	-.03
W	ST1-ST2 E	.60	.27	.23	.18	.08	.04	.00	-.07	-.02	.02
e	ST2-ST3 E	.43	.21	.16	.07	-.03	-.02	-.01	-.02	-.03	-.03
e											
k	ST3-ST5 N	.29	.15	.09	.01	-.05	-.05	-.05	-.02	-.02	.04
	ST4-ST5 N	.70	.57	.50	.42	.28	.24	.18	-.01	-.12	-.07
1	ST3-ST4 N	.42	.19	.13	.08	.01	.00	.00	.03	.05	.07
	ST3-ST5 E	.28	.03	.01	-.01	.01	-.00	.01	.05	.04	.03
	ST4-ST5 E	.61	.32	.25	.21	.16	.12	.05	-.07	-.05	-.03
	ST3-ST4 E	.37	.14	.07	-.02	-.08	-.07	-.05	.00	.01	-.01
	ST1-ST3 N	.79	.46		.37		.34		.24	.09	
	ST1-ST2 N	.79	.53		.41		.37		.17	.00	
	ST2-ST3 N	.62	.41		.32		.29		.14	.05	
	ST1-ST3 E	.74	.49		.44		.41		.22	.09	
W	ST1-ST2 E	.59	.41		.32		.21		.08	-.01	
e	ST2-ST3 E	.49	.24		.20		.17		.13	.06	
e											
k	ST3-ST5 N	.80	.48		.38		.36		.26	.20	
	ST4-ST5 N	.96	.80		.79		.69		.45	.26	
2	ST3-ST4 N	.77	.47		.36		.34		.15	.05	
	ST3-ST5 E	.50	.24		.26		.29		.22	.14	
	ST4-ST5 E	.86	.80		.74		.56		.22	-.03	
	ST3-ST4 E	.39	.19		.21		.22		.14	.04	

In the ideal case the longitudinal correlations (ST1-ST3 N, ST1-ST2 N ST3-ST5 E, ST4-ST5 E, ST3-ST4 E) were supposed to form an exponential function, but that showed up to be far from reality, with the highest correlation not always found at the smallest distance. This can eventually be explained in terms of small differences in the depths at which the meters were placed.

7.4. Time series analysis

A time series, i.e. a realisation of a stochastic process, can be described conveniently using the Box-Jenkins method (Box and Jenkins, 1976). A complicated signal is modelled using only a few parameters and a "white noise" process (independent normally distributed random shocks) like this:

$$Z_t = A_t + \theta_1 A_{t-1} + \theta_2 A_{t-2} + \ldots\ldots$$

where Z_t is the process at time t, A_t the white noise process at different time steps, and θ_i real numbers. This representation is called moving average since the process is a kind of an average of the present and earlier random shocks. By recursively solving this equation for the A_t's, Z_t can be written as a linear combination of earlier values of the process itself:

$$Z_{t-1} = A_{t-1} + \theta_1 A_{t-2} + \theta_2 A_{t-3} + \ldots\ldots$$

and thereby

$$A_{t-1} = Z_{t-1} - \theta_1 A_{t-2} - \theta_2 A_{t-3} \ldots\ldots$$

and so on. This gives after some calculations,

$$Z_t = \phi_1 Z_{t-1} + \phi_2 Z_{t-2} + \ldots\ldots + A_t$$

where ϕ_i can be found from the θ_i's

This last representation is called an autoregressive process, since it depends on earlier values of the process itself. If θ_i is zero after q time steps we talk about a moving average process of order q (a MA(q)-model), and similarly, if ϕ_i equals zero after p time steps, we call it an autoregressive process of order p (an AR(p)model). These two models can be combined to form an ARMA(p,q)-model. For instance, an ARMA(1,1)-model can be described as follows:

Appendix

$$z_t = \phi_1 z_{t-1} + \theta_1 A_{t-1} + A_t$$

An example of this can be found in Spliid et al. (1981) where the north component of the current velocity sampled each 10 minutes at a place in the Sound is expressed as

$$z_t = 0.96\ z_{t-1} - 0.64\ A_{t-1} + A_t$$

One of the advantages in using ARMA-models is that seasonal variations (24h, 7 days, 12 months or whatever it might be) are easy to incorporate. An example is the monthly catch of the American lobster 1967-1981 (Fogarty, 1984), which has been estimated at

$$z_t = \theta_1 A_{t-1} + \theta_{12} A_{t-12} + \theta_1 \theta_{12} A_{t-13} + A_t$$

$$= 0.4317\ A_{t-1} - 0.3417\ A_{t-12} - 0.1475\ A_{t-13} + A_t$$

thus only two parameters are used, and the model fit the data very well, and gives an easy tool in forecasting the future catch figures.

This type of statistical method is the one used in the Barsebäck current study, designed to get data to apply to a diffusion simulating model. This model (as described below) simulates the simultaneous movement of three particles, where the correlation is responsible for that between the A_t's to each particle, and thereby for the change in distance between particles. The ARMA-model will describe the movement of each particles, i.e. the model is equal for each particle, but the input (A_t) differs.

The analysis on the current measurements suggest an AR(2)-model, i.e. $p = 2$ and $q = 0$, leading to an autoregressive model. ARMA-processes have been estimated on many different data: from each

station, each week after different filtrations, on shorter periods, etc., as seen elsewhere.

Using the aforementioned cut-off frequency of 7 hours and estimating a model on the data from Station 1 on the first week, the following model was found for the north component, u, and east component, v:

$$u: Z_t = 0.59\ Z_{t-1} + 0.05\ Z_{t-2} + A_t$$
$$v: Z_t = 0.37\ Z_{t-1} + 0.12\ Z_{t-2} + A_t$$

using independent estimates for the two series.

This estimate, which can be the leading scheme in the simulation model, has been checked in several ways. The original and theoretical autocorrelations have been compared, as well as the original and theoretical spectra. The residuals from the estimates have been checked for "white-ness", etc. All tests showed satisfactory results, thus confirming the reliability of the estimated model.

Other models (other values for p and q) have also been estimated, but none gave a significantly better description of the data. For other stations and other periods similar models have been found.

Using all this information the simulation model can be constructed, where three particles released at the same time are looked upon. They are imposed by the slowly varying mean current (the deterministic part) and by the AR(2)-processes for N and E direction. Their correlation is expressed in that between the input (the A_t's) to the model, and this correlation is changed with time according to their relative orientation and separation. For simplicity an exponential correlation function is used for the longitudinal correlation with the characteristic length scale λ = 900 m. Details about the structure of the model can be found in (Spliid et al. 1981).

Appendix

Simulating the movement of a single particle, the two independent models for north and east velocity are used. Thus a picture like the one below can be seen.

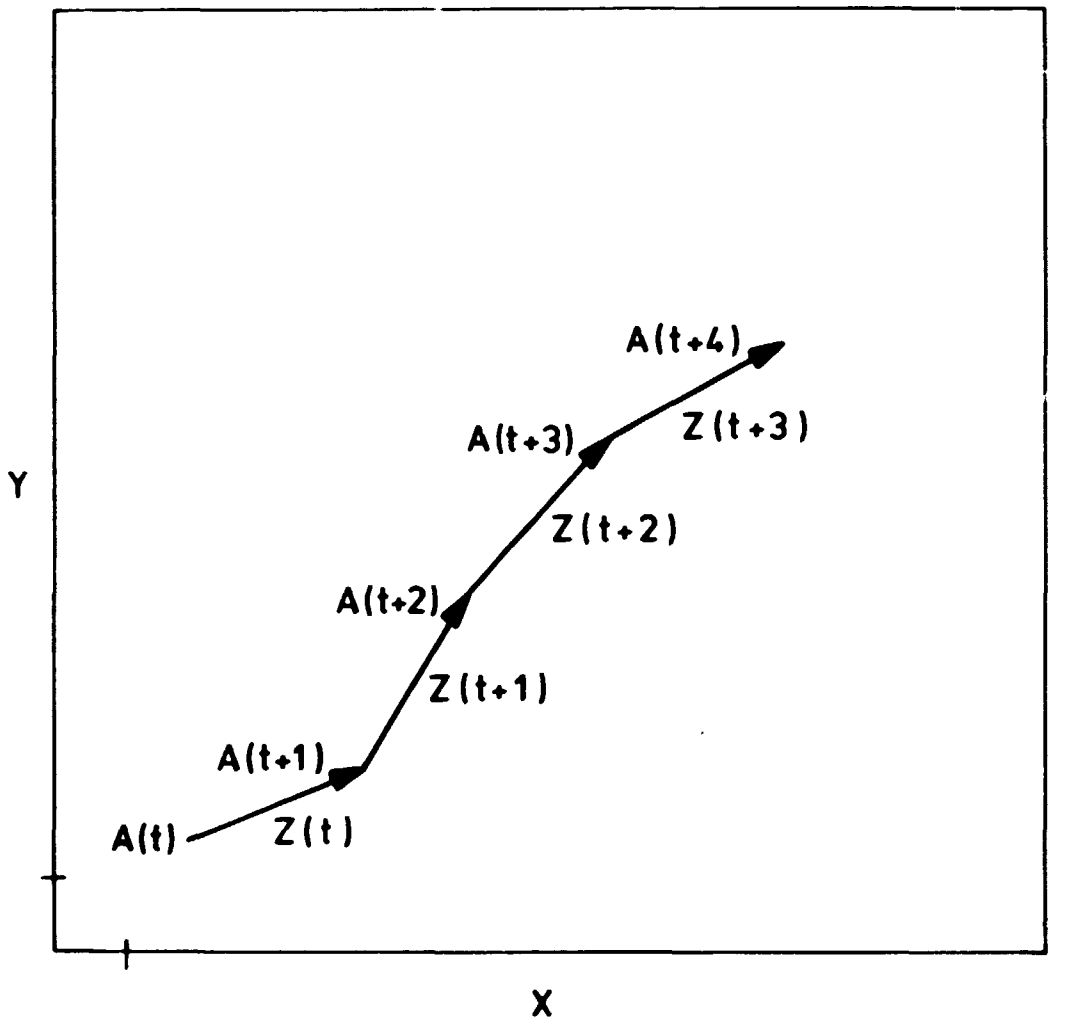

Fig. 7.5. Simulation of the movement of a single particle.

Adding a new particle creates a band on the random shocks, which now are correlated to a degree dependent on the distance between the particles. As written above, particles close together result in similar movements, far from each other means independent movements. For two particles an example is seen below.

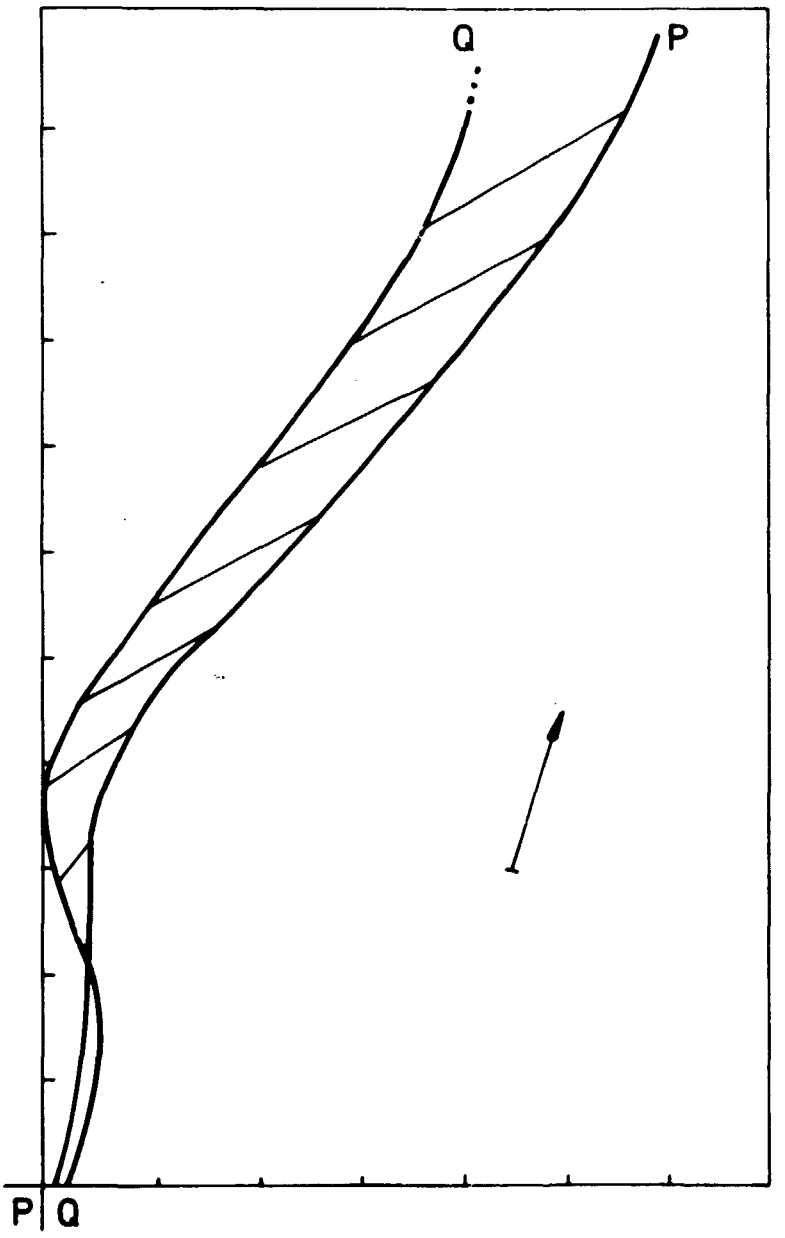

Fig. 7.6. Simulation of the simultaneous movements of two particles.

Appendix

If three particles are used the sum of the squared main axes of the smallest ellipse through the three points is a measure of the area covered or, equivalently the dilution. These ellipses can be drawn, for instance, for every 3 hours, as seen in the figure.

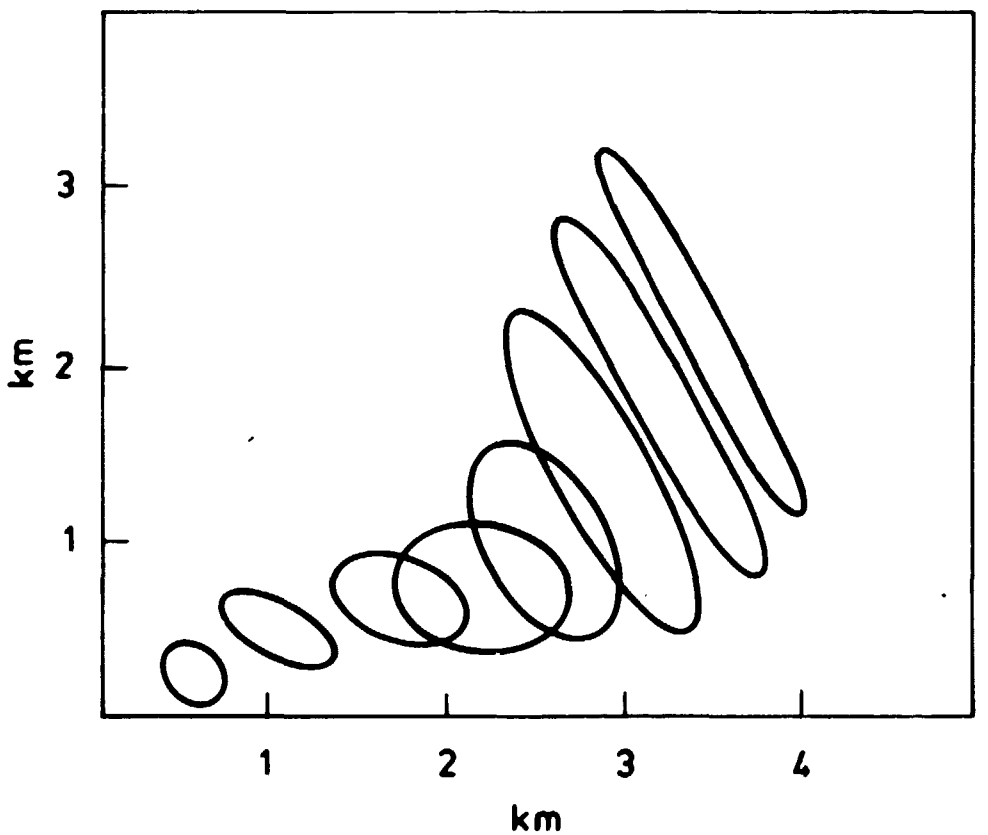

Fig. 7.7. Simulation example with the three-moving-points model. The ellipses are the smallest possible through three simultaneously moving points. The time interval between ellipses is 3 hours.

Appendix

Table 2. The dilution for different simulations (a-h) and the estimated average dilution. Each simulation reflects the average of 9 single experiments, and the dilution is expressed relative to that at time t=o.

	4 hr	8 hr	12 hr	16 hr	20 hr	24 hr
a)	3	9	24	40	44	50
b)	1	3	5	10	19	23
c)	4	9	13	20	31	47
d)	3	8	10	12	14	22
e)	3	8	13	17	22	29
f)	2	4	6	9	26	19
g)	2	7	24	22	28	33
g)	3	4	5	7	13	19
total	3	7	11	17	23	30

This type of model can give a picture of possible dilution processes for the area, and it can give a detailed description of the mean dilution as a function of time. The results from the model can be verified by the bioindicator measurements reported elsewhere. In Table 2 the dilution as a function of time is given for different simulations, all covering a time period 0-24 hours. A regession line can be found for the mean values for the dilution

$$D = \alpha \cdot T^{-1.3}$$

where T is the time in hours and α is a constant. This is in remarkable agreement with the Fucus results where the dependence of distance of ^{60}Co concentration as a function of distance from Barsebäck is found to be proportional to $X^{-1.4}$ (X in km).

Appendix

7.5. Conclusion

From the investigation of transport - diffusion models it became obvious that if a model based only on current measurements could be constructed, it would result in a much easier data collecting procedure than the usual method involving tracer experiments. Therefore, the current-measuring programme near Barsebäck was set up, and an analysis of the results showed that after eliminating periods longer than 7 hours no significant correlation could be found at 900 meters. This information is used in the simulation model, which also is based on autoregressive models estimated on the current velocity data. The model gives a discription of the dilution in the area north of Barsebäck and the results have been verified by bioindicator measurements.

Appendix

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Abstract

This project describes the application of bioindicator systems intended for the measurement of the low level radioactive contamination around nuclear installations. The system has been applied around the Swedish and Finnish nuclear power plants and has, furthermore, been used in a study of the dispersion of the effluents from the British nuclear reprocessing plant, Sellafield. The doses to man from these installations have been calculated and compared with the natural background radiation received from the consumption of marine fish.

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